RADIOACTIVE AIR EMISSIONS FROM CANDU NUCLEAR POWER PLANTS

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D R A F T

SUMMARY AND CONCLUSIONS

This paper is a sequel to a report entitled "Radioactive Liquid Wastes from Steam-Electric Generating Stations" which was prepared for the Radioactivity Working Group of the Task Force on Steam-Electric Power Generation.

The paper examines radioactive airborne emissions from CANDU nuclear powered generating stations and describes available technologies for the control and treatment of atmospheric emissions.

Tritium and carbon-14 are the two radionuclides of primary interest in airborne effluents from CANDU stations. Both have residence times in the atmosphere and hydrosphere sufficiently long enough for transport processes to distribute them globally. Other fission and activation products occurring in airborne effluents include the noble gases, iodine -131 and particulates e.g. strontium -90, calcium -137, cobalt -60 etc.

With the exception of carbon -14, radioactive airborne emissions from CANDU stations have historically been below one percent (1%) of the current Canadian regulatory Derived Emission Limits (DELs), promulgated by the Atomic Energy Control Board. A fixed DEL has not been established for carbon -14 although a Provisional Derived Emission Limit (PDEL) is available.

Annual tritium emissions from CANDU stations have varied from 110 $TBq.a^{-1}$ for Bruce 'A' Nuclear Generating Station (NGS) in 1976 to 1600 $TBq.a^{-1}$ at Pickering 'A' NGS in 1977. Noble gas emissions vary from 22 $TBq\text{-}MeV.a^{-1}$ at Bruce NGS 'A' in 1976 to 5000 $TBq.\text{-}MeV.a^{-1}$ at Douglas Point NGS in 1973. Particulate emissions range from non-detectable at Nuclear Power Demonstration (NPD) NGS in 1974 to 1.3 $TBq.a^{-1}$ at Pickering NGS 'A' in 1974. Iodine -131 emissions vary from non-detectable at NPD NGS in 1975 to 4.9×10^{-3} $TBq.a^{-1}$ at Pickering NGS 'A' in 1972. Measurements of average weekly carbon -14 emissions range from 1.3 Ci/week Pickering NGS 'A' to 16.4 Ci/week at Pickering NGS 'A' for one year study periods during 1981 and 1982. The latter emission represents 0.93% of the PDEL for carbon -14.

Tritium concentrations in heavy water circuits increase with time until an equilibrium concentration is reached when the rate of production equals the decay rate plus the rate of change of tritium concentrations due to heavy water losses. The rate of increase of tritium is greater in the moderator than in the primary heat transport heavy water. Potential sources of tritium emissions are leaks and spills from the moderator system and the primary heat transport system.

Carbon -14 is mostly produced in the moderator heavy water and the coolant heavy water, 95% being produced in the moderator.

Design and operational approaches that reduce the quantity and concentrations of airborne emissions from CANDU stations include containment, controlled venting and treatment.

Tritium emissions can be reduced by the minimization of heavy water leaks and spills, the collection, upgrading and re-use of heavy water and the use of a tritium removal system applied to tritiated heavy water circuits. However, at present the principal component of airborne emissions control is the placement of most of the heavy water systems within the reactor containment structure which is fitted with a closed cycle heavy water vapour recovery system.

Most carbon -14 emissions would be expected to travel from the source into containment areas, recirculating vapour recovery systems and then via a purge stream to the reactor building exhaust and stack. At present, sampling data is required only to satisfy compliance requirements since control monitoring of C-14 emissions is not required. No treatment is currently being carried out.

A central noble gas off-gas treatment system is installed at each major station to reduce emissions associated with fuel failures. The system uses ambient temperature charcoal delay beds.

The exhaust from contaminated ventilation systems passes through a filtration unit consisting of a prefilter, HEPA filter and a TEDA impregnated charcoal bed prior to stack monitoring for particulate, I-131 noble gas and tritium.

An attempt is made to evaluate the effectiveness of alternative treatment technologies in reducing radioactive emissions from CANDU stations in accordance with the principle of Best Practicable Technology (BPT), the control philosophy currently favoured by the Environmental Protection Service (EPS) of Environment Canada (EC).

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1 INTRODUCTION

This paper examines radioactive air emissions from nuclear generating stations and describes alternative technologies for the control of gaseous radioactive emissons. The effects of radionuclide emissions on the aquatic environment or on human beings are not addressed, rather, emphasis is placed on technical aspects of the various systems that can be used to reduce such emissions from power stations.

The CANDU reactor system, in use by Ontario Hydro, employs natural uranium as fuel together with a pressurized heavy water (D_2O) heat transport system and an independent, low pressure heavy water moderator system.

Four nuclides, tritium, carbon-14, krypton-85 and iodine-129, were identified in a recent report of the Nuclear Energy Agency (the Polvani Report (1)) as being the nuclides in effluents most likely to be significant for the exposure of large populations from installations associated with the nuclear fuel cycle. Tritium and carbon-14, which have half-lives of 12.3 a and 5730 a respectively, are both produced and emitted by CANDU reactors and have residence times in the atmosphere and hydrosphere sufficient for transport processes to distribute them globally.

2 CHARACTERIZATION OF GASEOUS EFFLUENTS FROM CANDU NUCLEAR POWER STATIONS

2.1 General Description of Sources and Radioactivity Emissions

2.1.1 Sources of Radioactivity. The CANDU reactor, used by Ontario Hydro, employs natural uranium in the form of uranium dioxide as fuel together with a pressurized heavy water (D_2O) heat transport system and an independent, low pressure heavy water moderator system.

A partial listing of various nuclear reactions associated with CANDU reactors is shown in Table A.1, Appendix A. The sources of radioactivity in gaseous wastes from nuclear stations are fission products (from the fission process in the reactor), activation products and activated corrosion products (due to the conversion of an element to a radioactive isotope by addition of a neutron).

(i) Fuel Fission Products

In the reactor each fissoning nucleus produces two heavy nuclei and an average of between two and three high energy neutrons. The uranium isotopes form approximately 80 chains of fission products which result in over 200 individual fission products (2). Some fuel fission products and their half-lives are listed in Table B.2 (1), Appendix B.

These fission products may escape from the fuel bundles in the reactor or in the irradiated fuel bay. However, fuel sheath failures or defects and imperfections in fuel cladding are few in number and increasingly uncommon. It is more difficult for fission products to escape in particulate form due to fuel sheath defects than as gaseous fission products. A number of fission product noble gases decay to particulate material. Gaseous fission products may also escape by molecular permeation through an intact fuel sheath.

(ii) Activation Products

Elements in the materials of construction of the various reactor systems may become activated by neutron bombardment. Most of this material remains fixed inside the reactor. However, any material that dissolves or exists as particulate corrosion products in the Primary Heat Transport (PHT) system, will pass through the reactor core and may become activated. This will also occur in the Moderator circuit but to a lesser extent.

A partial listing of activated corrosion products, together with their parent isotopes is presented in Table B.3 (2), Appendix B.

Heavy water in the moderator and primary heat transport systems also becomes activated. The activation of deuterium produces tritium, which exists in the form of tritiated heavy water (DTO) or, more commonly, tritiated water (HTO).

Preliminary tabulations of fission and activation products of environmental interest for Heavy Water Reactors (HWRs) are presented in Tables 1 and 2 respectively.

2.1.2 Emissions from Nuclear Stations. Most radioactive materials formed within the fuel elements are retained in these elements. Most radionuclides which diffuse into or are formed within reactor coolant systems are retained within those systems. However, radionuclides may reach the environment through gaseous, liquid or solid pathways.

Traditionally, the reporting of radioactive airborne emissions has been expressed in Curies/annum (Ci/a) for tritium, particulates and radioiodine and in Ci-Mev/a for the noble gases. More recently the S.I. unit of radioactivity, the bequerel (Bq) has replaced the Curie. These emissions are also expressed as a percentage of the Derived Emission Limits (DELs) which are site specific limits derived by Ontario Hydro.

The annual airborne emissions of tritium, iodine-131, noble gases and particulates from 1972 to 1980 are presented in Figures 1 to 16 for the 2000 MWe Pickering 'A' Nuclear Generating Station (PNGS 'A'), 200 MWe Douglas Point NGS (DPNGS), 22 MWe Rolphton Nuclear Plant Demonstration NGS (NPD NGS) and the 3000 MWe Bruce 'A' NGS (BNGS 'A') (5). Monthly data for Pickering and Bruce from 1977 to 1982 are presented in Figures 17 to 23 (6).

Annual tritium emissions from CANDU stations vary from 110 $TBq.a^{-1}$ for BNGS 'A' in 1976 to 1600 $TBq.a^{-1}$ at PNGS 'A' in 1977. Noble gas emissions vary from 22 $TBq\text{-}MeV.a^{-1}$ at BNGS 'A' in 1976 to 5000 $TBq\text{-}MeV.a^{-1}$ at DPNGS in 1973. Particulate emissions range from non-detectable at NPDNGS in 1974 to 1.3 $TBq.a^{-1}$ at PNGS 'A' in 1974. Iodine-131 emissions vary from non-detectable at NPD NGS in 1975 to 4.9×10^{-3} $TBq.a^{-1}$ at PNGS 'A' in 1972.

Carbon-14 emissions are indicated in Table 3 for the above mentioned plants and graphical representations for Pickering and Bruce are shown in Figures 24 and 25 (7). Since carbon-14 has only been monitored in recent years, only two years of data are presented. A provisional derived emission limit (PDEL) is indicated for $^{14}CO_2$ emissions. Measurements of average weekly carbon-14 emissions range from 1.3 Ci/week at BNGS 'A' to 16.4 Ci/week at PNGS 'A' for one year study periods during 1981 and 1982. Emissions from Pickering NGS 'A' are greater than emissions from any of the other stations as a large proportion of the C-14 originates from the N_2 - filled annulus gas system. Peaks in

TABLE 1 SELECTED FISSION PRODUCTS FROM CANDU REACTORS (2)

Element	Symbol	Half-Life	Specific Activity (Ci/gram)	Element	Symbol	Half-Life	Specific Activity (Ci/gram)
Strontium	Sr^{89}	52d	2.822×10^4	Iodine	I^{131}	8.070 ± 0.009d	1.235×10^5
Strontium	Sr^{91}	9.67h	3.561×10^6	Iodine	I^{132}	2.3h	1.032×10^7
Strontium	Sr^{90}	28.1y	1.414×10^2	Iodine	I^{133}	20.9 ± 1h	1.127×10^6
Yttrium	Y^{90}	64h	544.1×10^3	Iodine	I^{135}	6.7h	3.465×10^6
Yttrium	Y^{91}	58.8d	2.440×10^4	Xenon	Xe^{133}	5.27d	1.863×10^5
Yttrium	Y^{92}	3.53h	9.650×10^6	Xenon	Xe^{135}	9.2h	2.523×10^6
				Cesium	Cs^{134}	2.05y	1.301×10^3
Yttrium	Y^{93}	10.2h	3.304×10^6	Cesium	Cs^{137}	30.2 ± 0.2y	8.633×10^1
Zirconium	Zr^{95}	65d	21.15×10^3	Barium	Ba^{137m}	2.55m	5.383×10^8
Zirconium	Zr^{97}	17h	1.901×10^6	Barium	Ba^{140}	12.8d	72.87×10^3
Niobium	Nb^{95}	35.15 ± 0.03d	3.911×10^4	Lanthanum	La^{140}	40.2 ± 0.1h	5.566×10^5
Niobium	Nb^{97}	72m	26.92×10^6	Lanthanum	La^{141}	3.9h	5.699×10^6
Molybdenum	Mo^{99}	66.69 ± 0.06h	4.747×10^5	Cerium	Ce^{141}	33d	2.806×10^4
Ruthenium	Ru^{103}	39.6d	3.202×10^4	Cerium	Ce^{143}	33h	6.641×10^5
Ruthenium	Ru^{106}	367d	3.357×10^3	Cerium	Ce^{144}	284.9 ± 0.8d	3.183×10^3
Rhodium	Rh^{106}	30s	3.548×10^9	Praseodymium	Pr^{144}	17.3m	75.48×10^6
Rhodium	Rh^{103m}	54.7m	3.338×10^7	Praseodymium	Pr^{143}	13.7d	66.65×10^3
Rhodium	Rh^{105}	35.9h	8.314×10^5	Neodymium	Nd^{147}	11.1d	80.03×10^3
Tellurium	Te^{132}	78h	34.4×10^3	Promethium	Pm^{147}	2.5y	9.728×10^2
				Promethium	Pm^{149}	53.1h	3.961×10^5

Note: Specific activities are calculated.
Half lives are from reference (3).

TABLE 2 SELECTED ACTIVATION PRODUCTION FROM CANDU REACTORS (4)

Element	Symbol	Half-Life	Specific Activity (Ci/gram)
Tritium	H^3	12.26y	9.720×10^3
Carbon-14	C^{14}	5730y	4.457
Argon-41	Ar^{41}	1.83h	4.177×10^7
Chromium-51	Cr^{51}	27.8d	9.21×10^4
Manganese-54	Mn^{54}	303d	7.981×10^3
Iron-59	Fe^{59}	45.1 ± 0.5d	4.907×10^4
Cobalt-58	Co^{58}	71.3d	3.158×10^4
Cobalt-60	Co^{60}	5.26 ± 0.01y	1.133×10^3
Copper-64	Cu^{64}	12.9h	3.796×10^6
Zinc-65	Zn^{65}	243.60 ± 0.01d	8.247×10^3
Zirconium-95	Zr^{95}	65d	2.115×10^4
Silver-110m	Ag^{110m}	253d	4.692×10^3
Antimony-124	Sb^{124}	60.3 ± 0.2 d	1.746×10^4
Wolfram-187 (Tungsten)	W^{187}	24h	6.983×10^5

Various sources of information used in addition to reference.
Half lives are from reference (3).

TABLE 3 AVERAGE C-14 EMISSONS (7)

Station	Predicted (Ci/WK)	Measured (Ci/WK)	% $^{14}CO_2$	% PDEL*
BNGS-A	16	1.3	78	0.09
PNGS-A	38	16.4	80	0.93
DPNGS	1.6	0.47	82	0.02
NPD	0.4	0.27	98	0.007

* Only $^{14}CO_2$ emissions are applied to the DELs

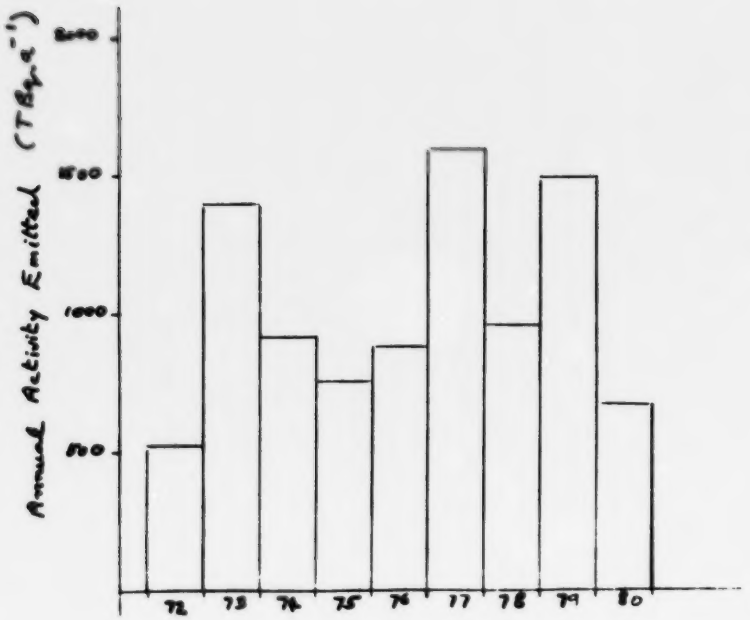

FIGURE 1 ANNUAL TRITIUM GASEOUS EMISSIONS FROM PICKERING NGS (5)

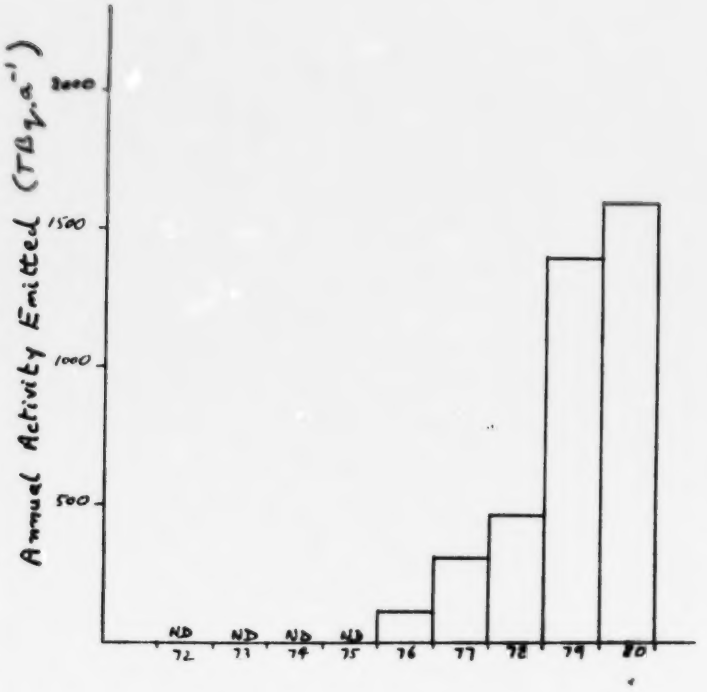

FIGURE 2 ANNUAL TRITIUM GASEOUS EMISSIONS FROM BRUCE NGS (5)

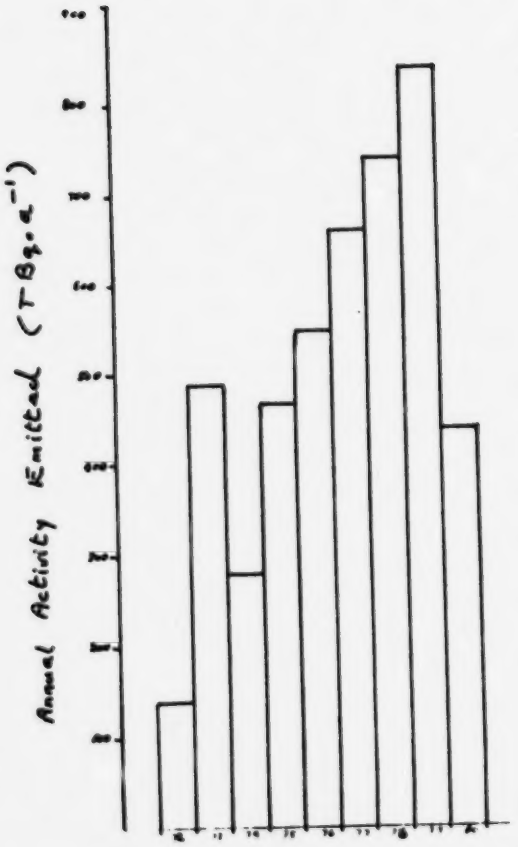

FIGURE 3 ANNUAL TRITIUM GASEOUS EMISSIONS FROM NPD NGS (5)

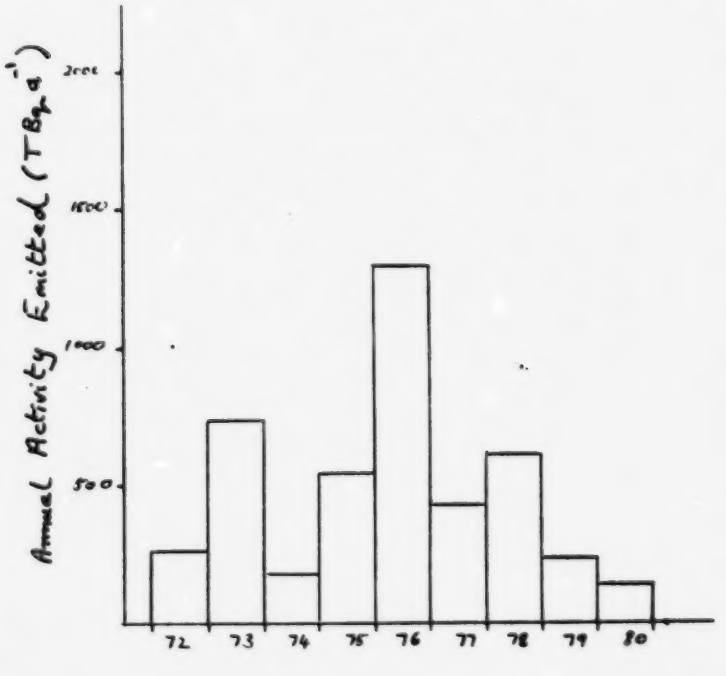

FIGURE 4 ANNUAL TRITIUM GASEOUS EMISSIONS FROM DOUGLAS POINT NGS (5)

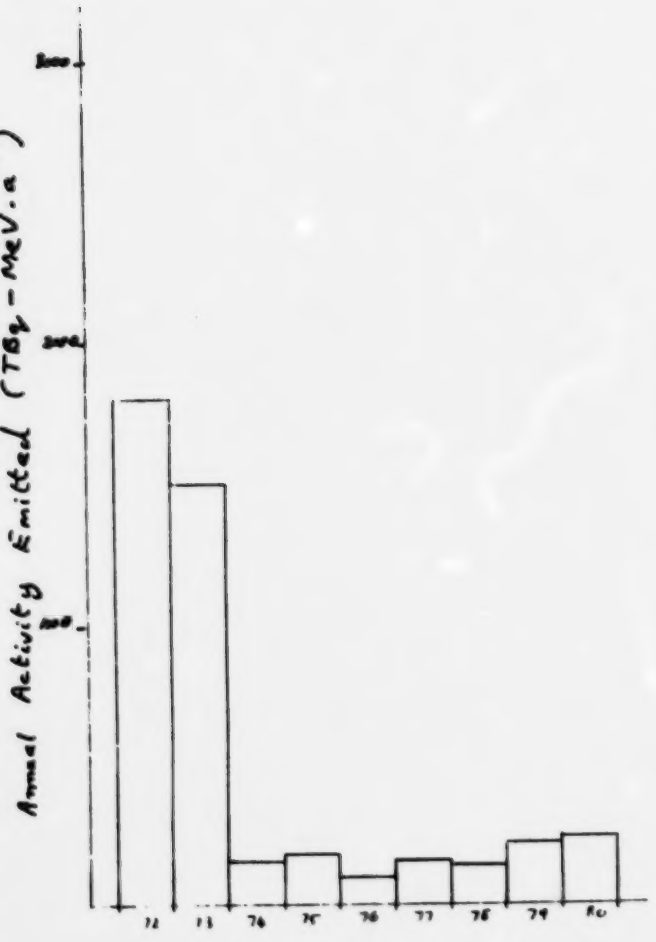

FIGURE 5 ANNUAL NOBLE GASES EMISSIONS FROM PICKERING NGS (5)

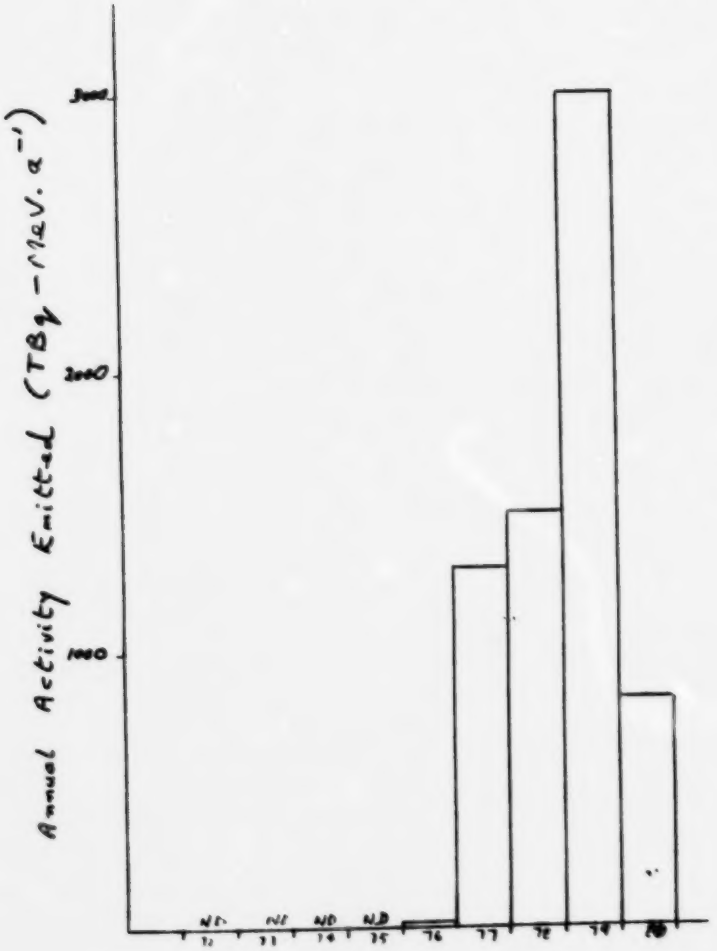

FIGURE 6 ANNUAL NOBLE GASES EMISSIONS FROM BRUCE NGS (5)

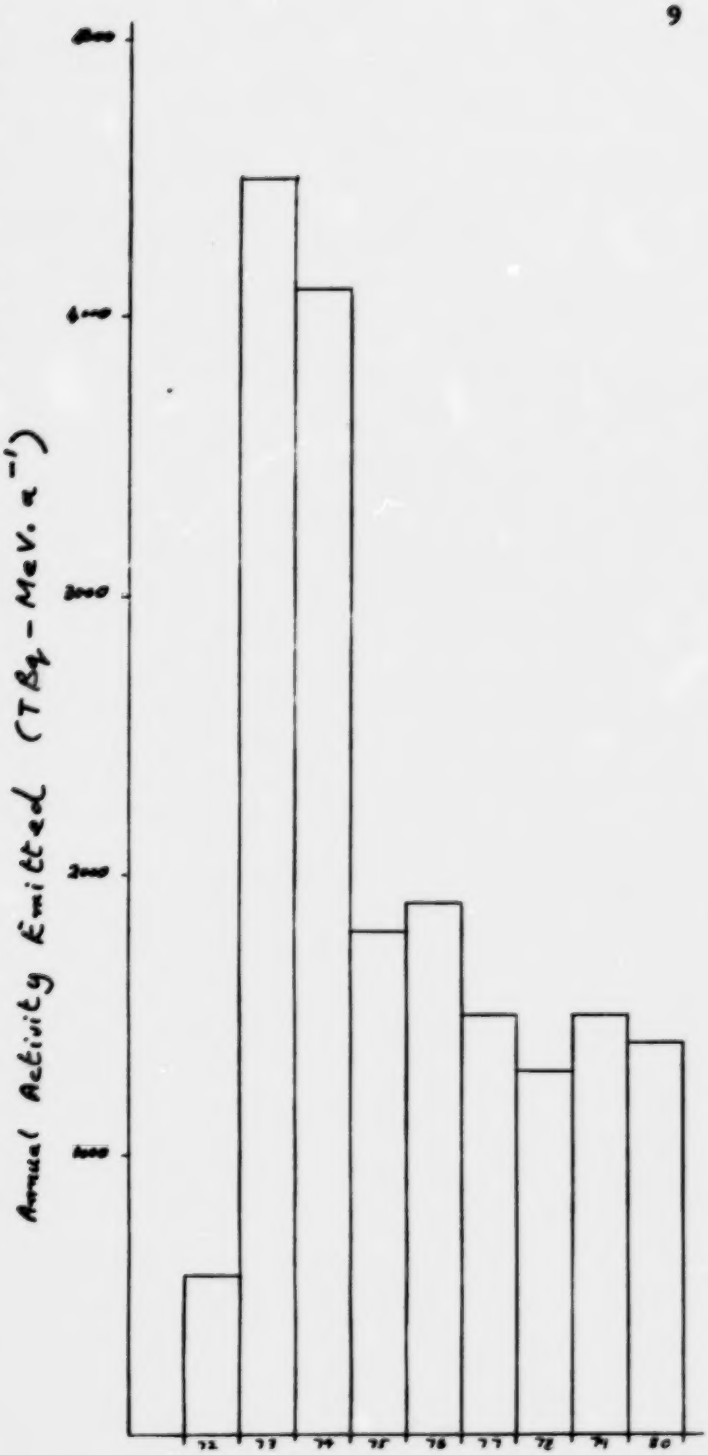

FIGURE 7 ANNUAL NOBLE GASES EMISSIONS FROM NPD NGS (5)

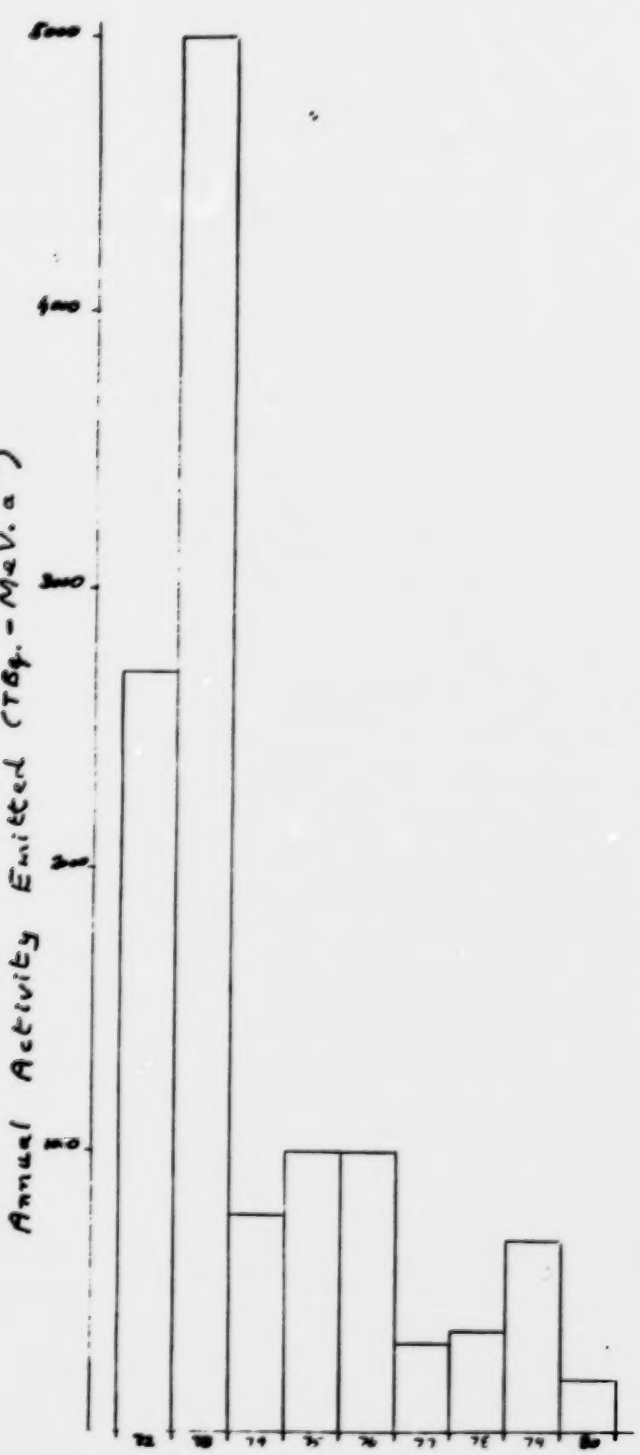

FIGURE 8 ANNUAL NOBLE GASES EMISSIONS FROM DOUGLAS POINT NGS (5)

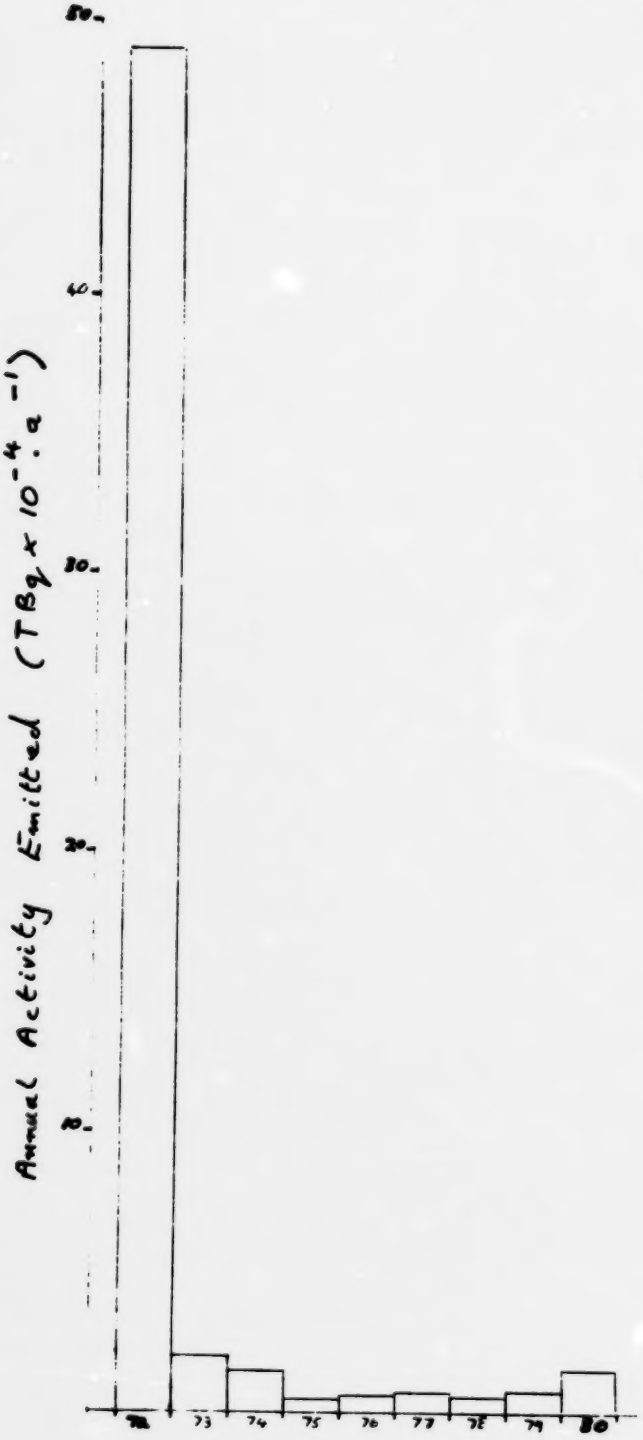

FIGURE 9 ANNUAL IODINE-131 GASEOUS EMISSIONS FROM PICKERING NGS (5)

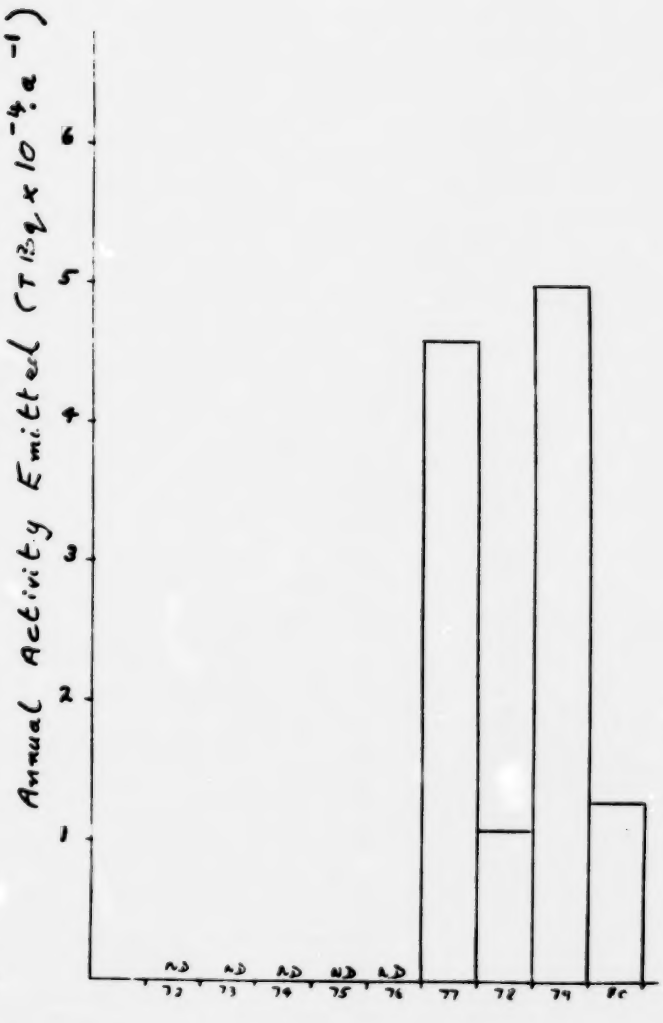

FIGURE 10 ANNUAL IODINE-131 GASEOUS EMISSIONS FROM BRUCE NGS (50

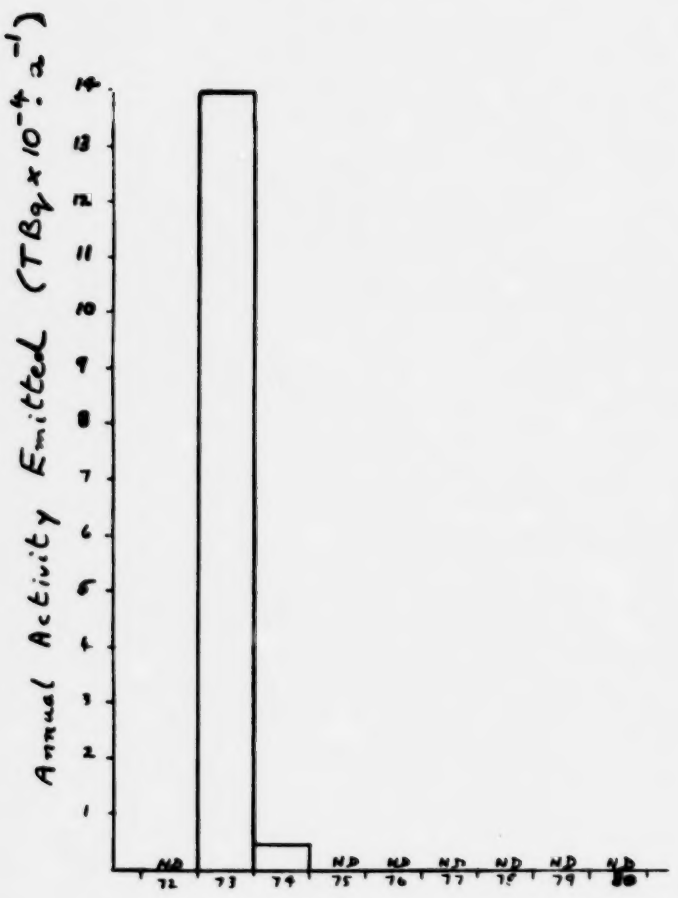

FIGURE 11 ANNUAL IODINE-131 GASEOUS EMISSIONS FROM NPD NGS (5)

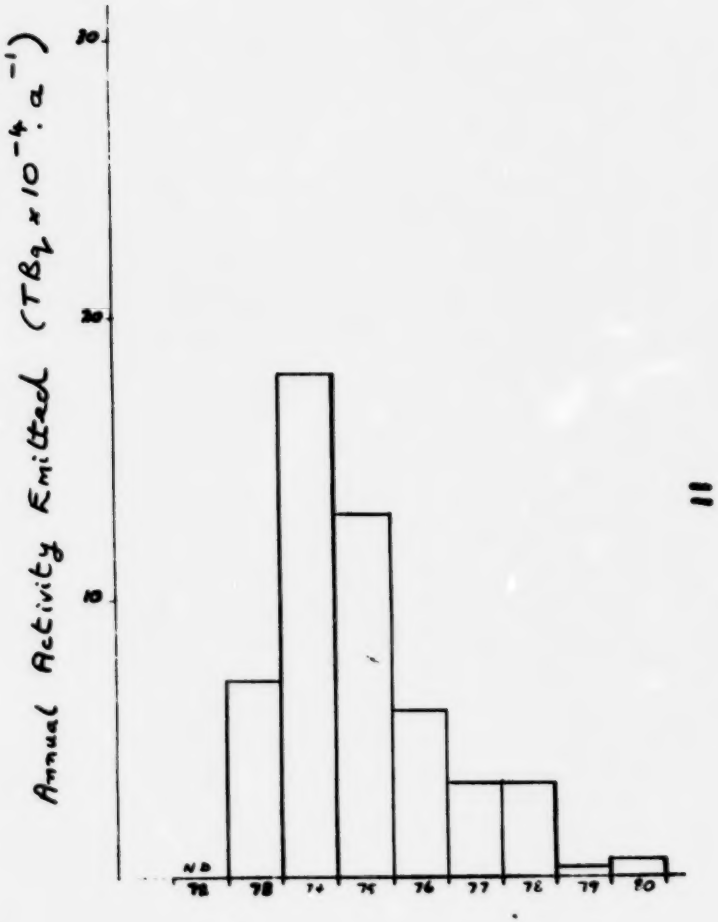

FIGURE 12 ANNUAL IODINE-131 GASEOUS EMISSIONS FROM DOUGLAS POINT NGS (5)

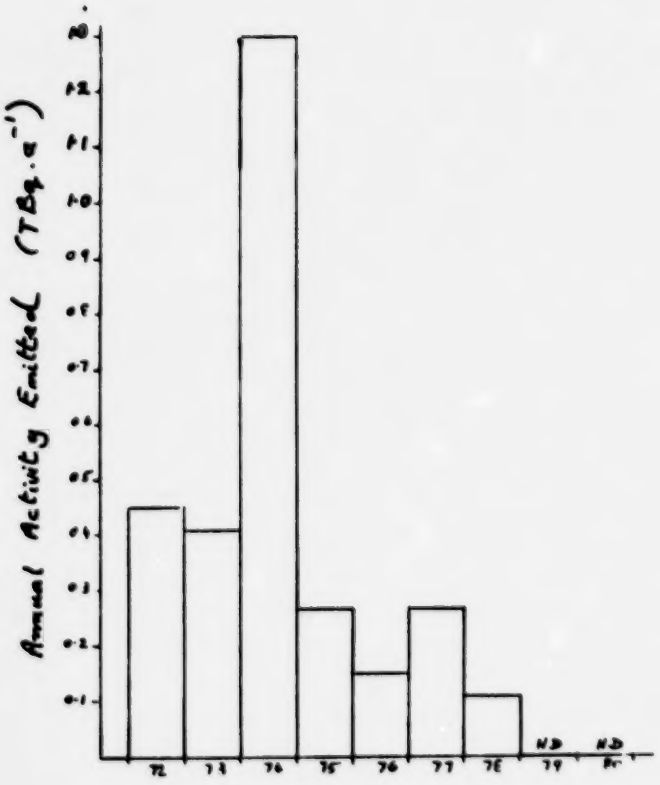

FIGURE 13 ANNUAL PARTICULATE GASEOUS EMISSIONS FROM PICKERING NGS (5)

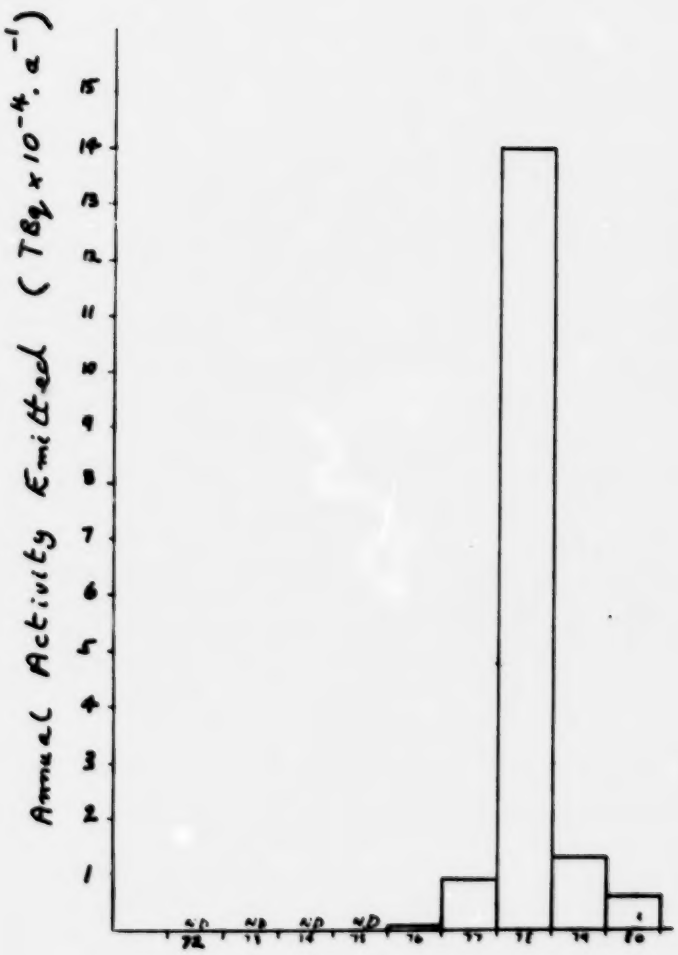

FIGURE 14 ANNUAL PARTICULATE GASEOUS EMISSIONS FROM BRUCE NGS (5)

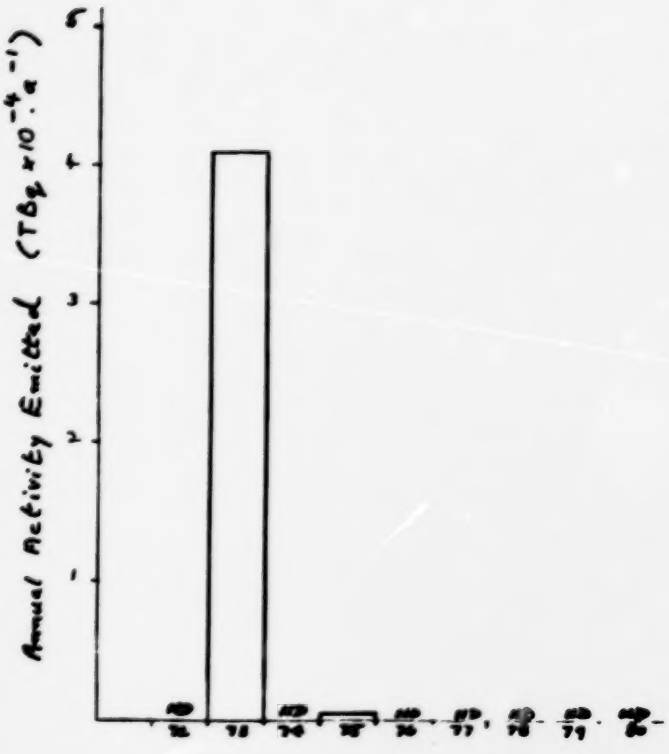

FIGURE 15 ANNUAL PARTICULATE GASEOUS EMISSIONS FROM NPD NGS (5)

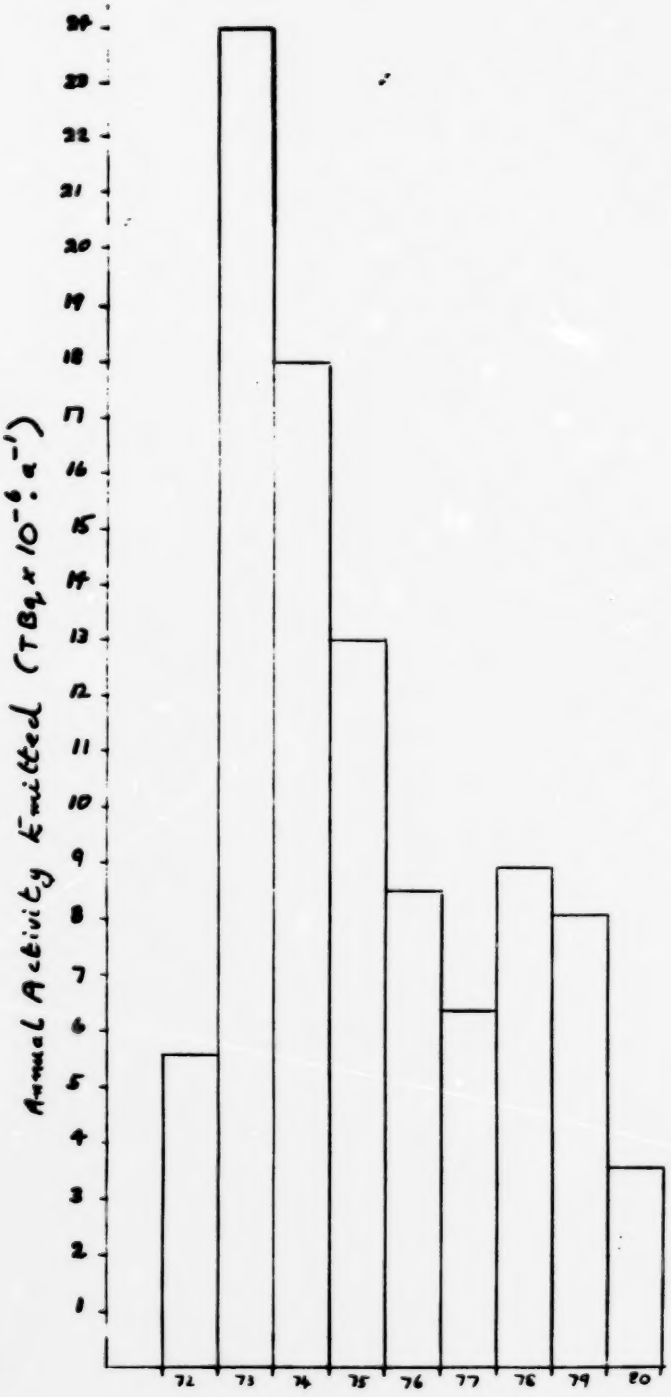

FIGURE 16 ANNUAL PARTICULATE GASEOUS EMISSIONS FROM DOUGLAS POINT NGS (5)

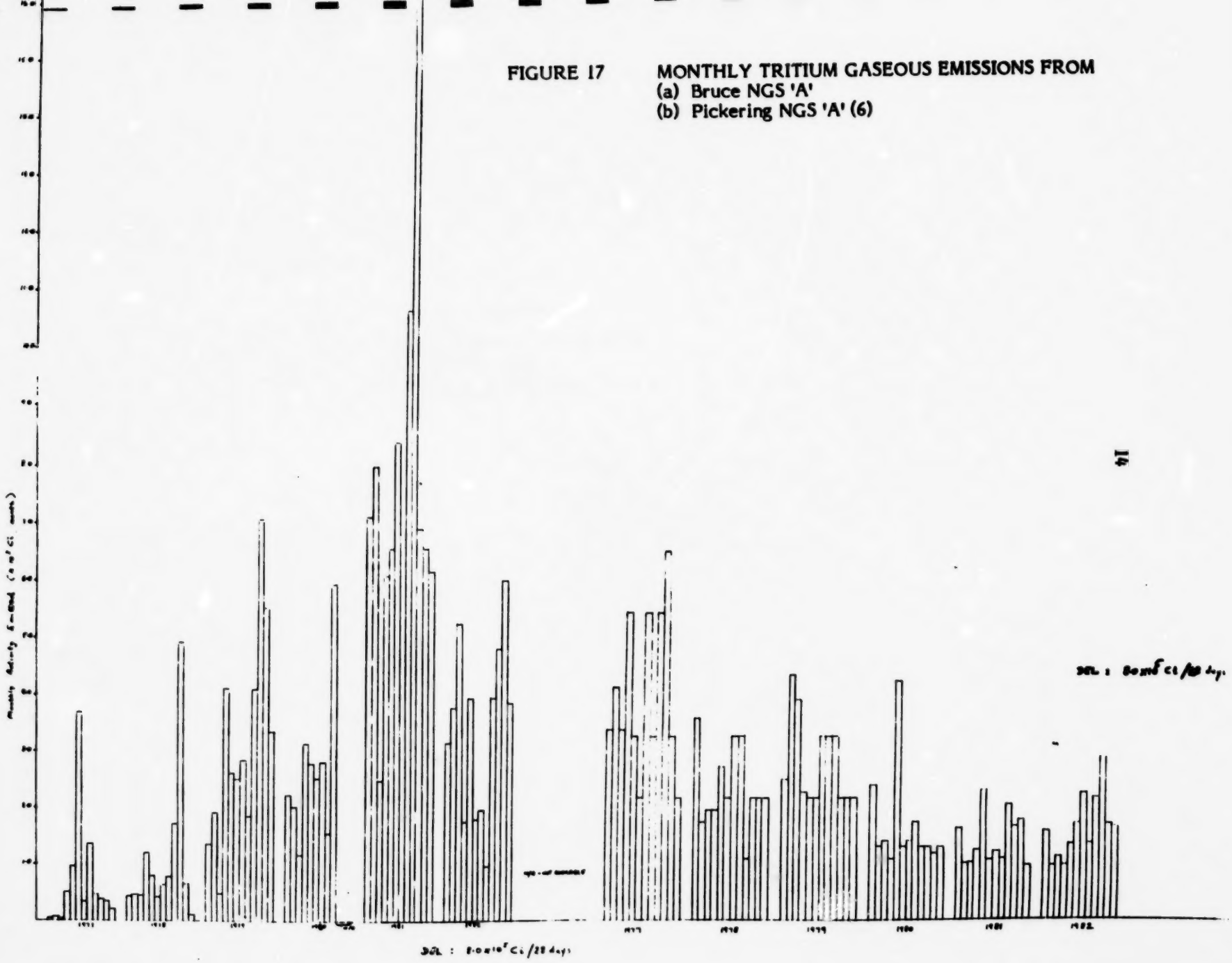

FIGURE 17 MONTHLY TRITIUM GASEOUS EMISSIONS FROM
(a) Bruce NGS 'A'
(b) Pickering NGS 'A' (6)

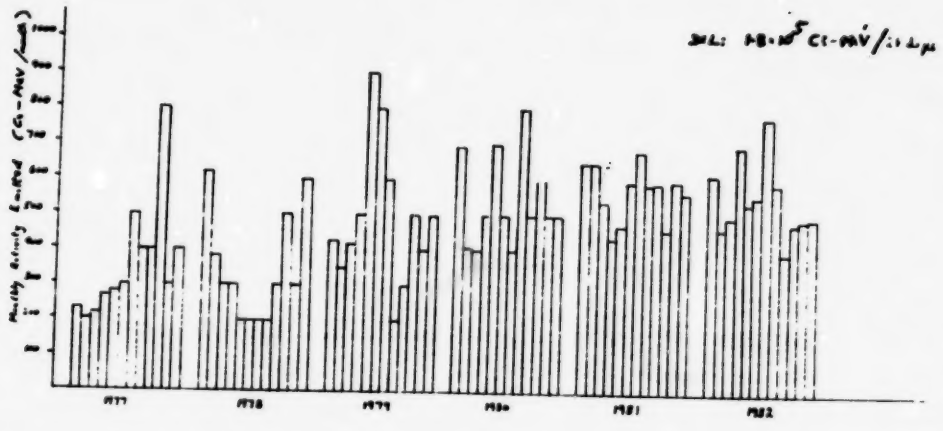

FIGURE 18 MONTHLY NOBLE GASES EMISSIONS FROM PICKERING NGS 'A' (6)

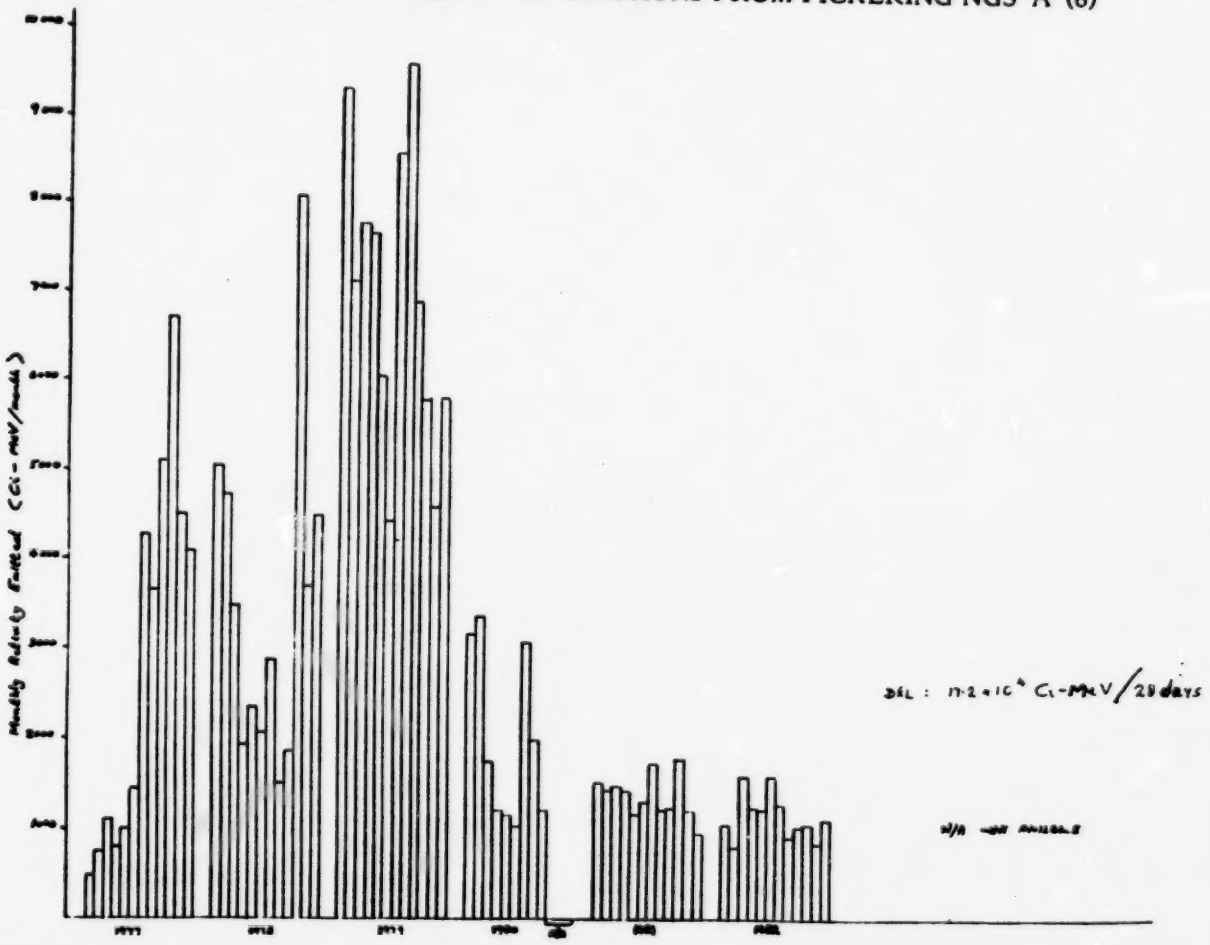

FIGURE 19 MONTHLY NOBLE GASES EMISSIONS FROM BRUCE NGS 'A' (6)

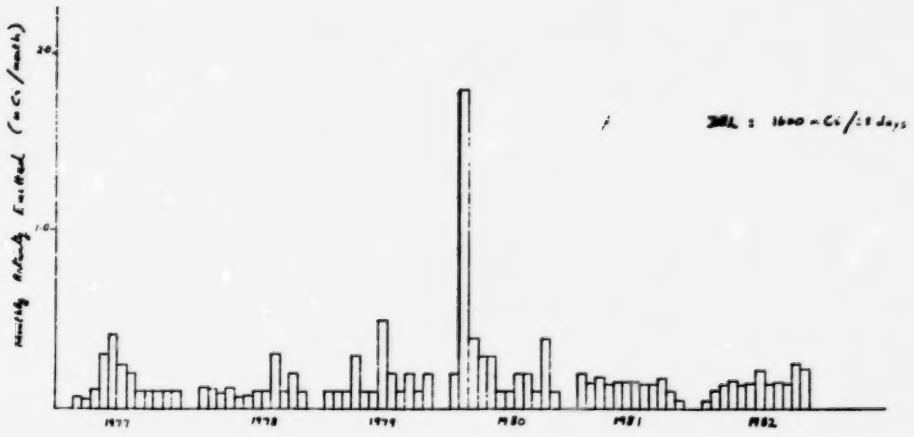

FIGURE 20 MONTHLY IODINE-131 EMISSIONS FROM PICKERING NGS 'A' (6)

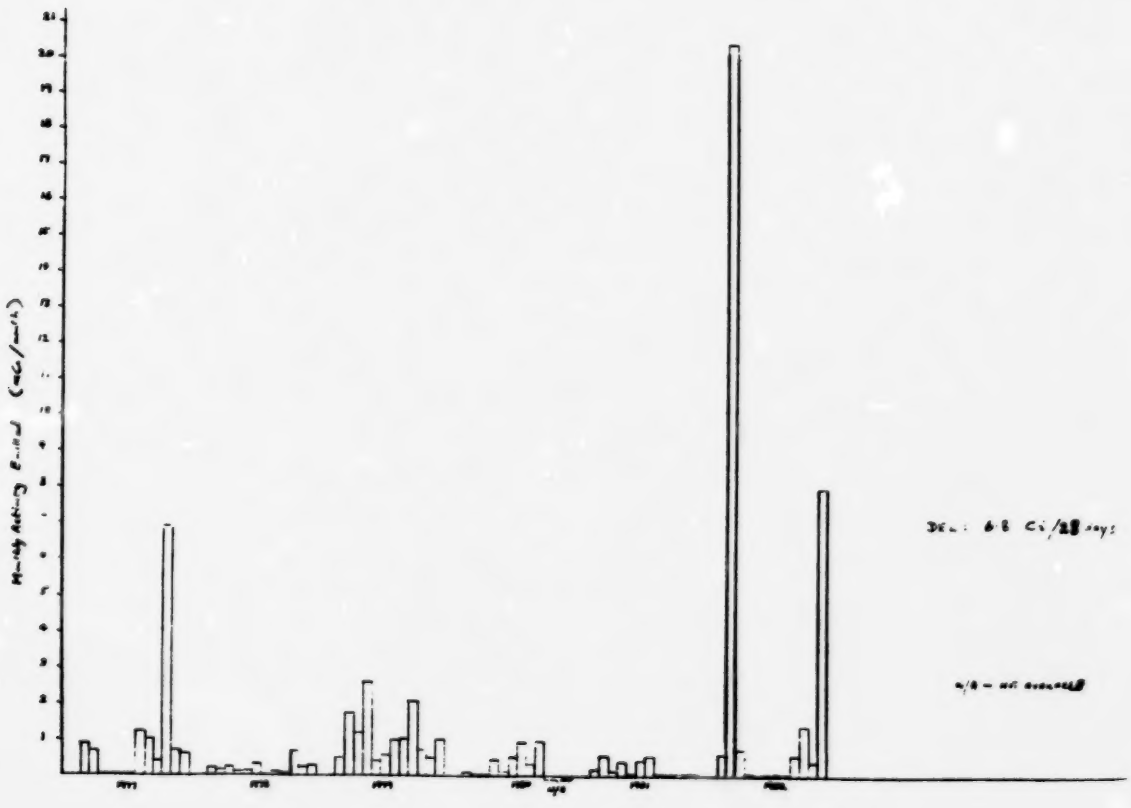

FIGURE 21 MONTHLY IODINE-131 EMISSIONS FROM BRUCE NGS 'A' (6)

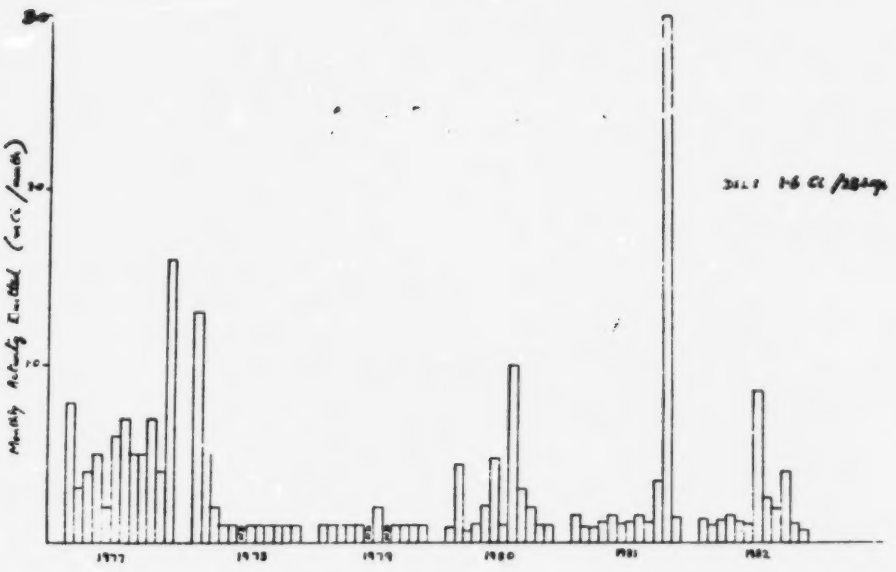

FIGURE 22 MONTHLY PARTICULATE GASEOUS EMISSIONS FROM PICKERING NGS 'A' (6)

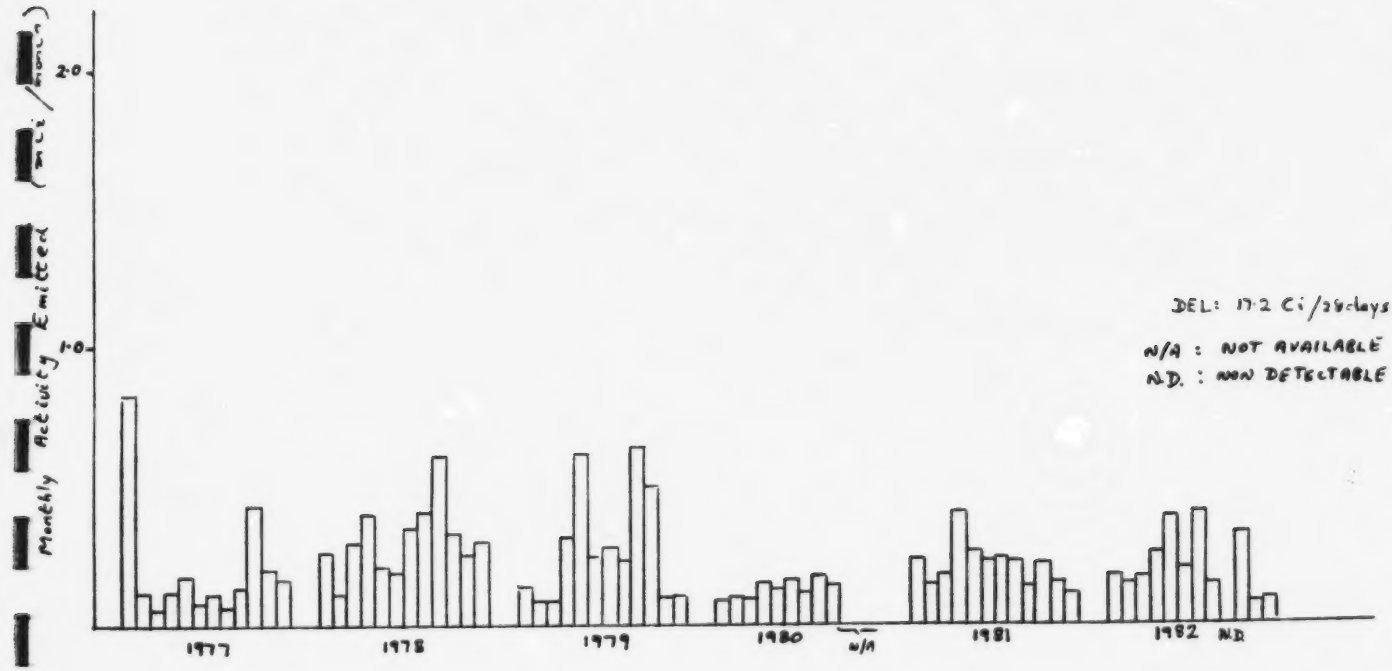

FIGURE 23 MONTHLY PARTICULATE GASEOUS EMISSIONS FROM BRUCE NGS 'A' (6)

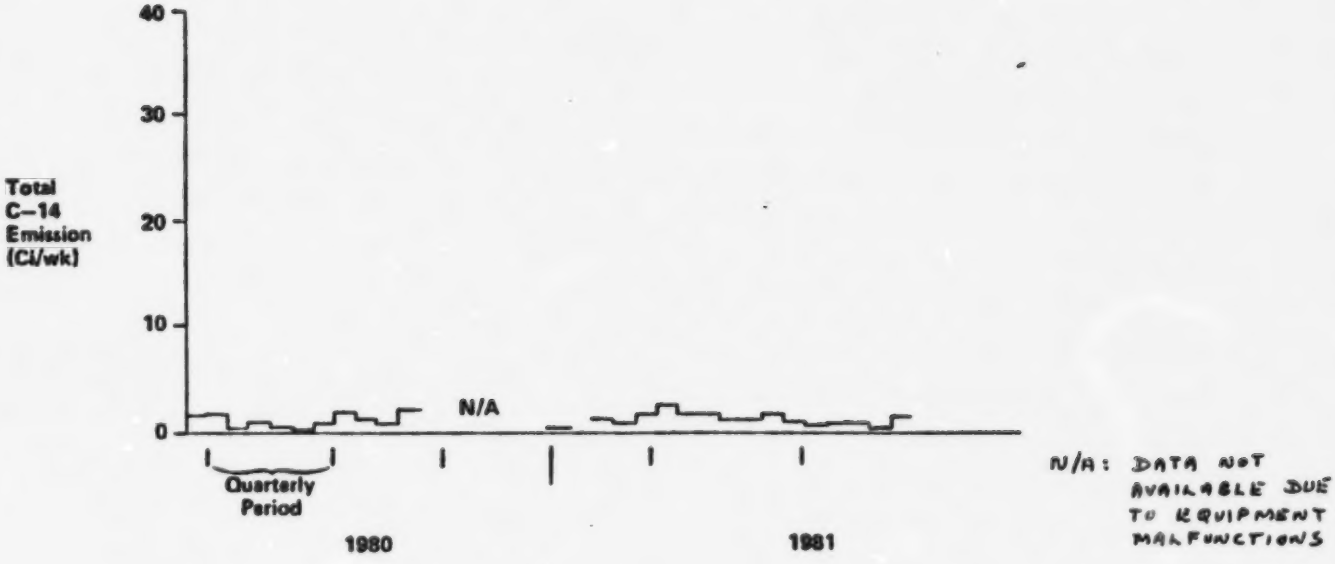

FIGURE 24 TOTAL EMISSIONS OF C-14 AT BNGS 'A' (7)

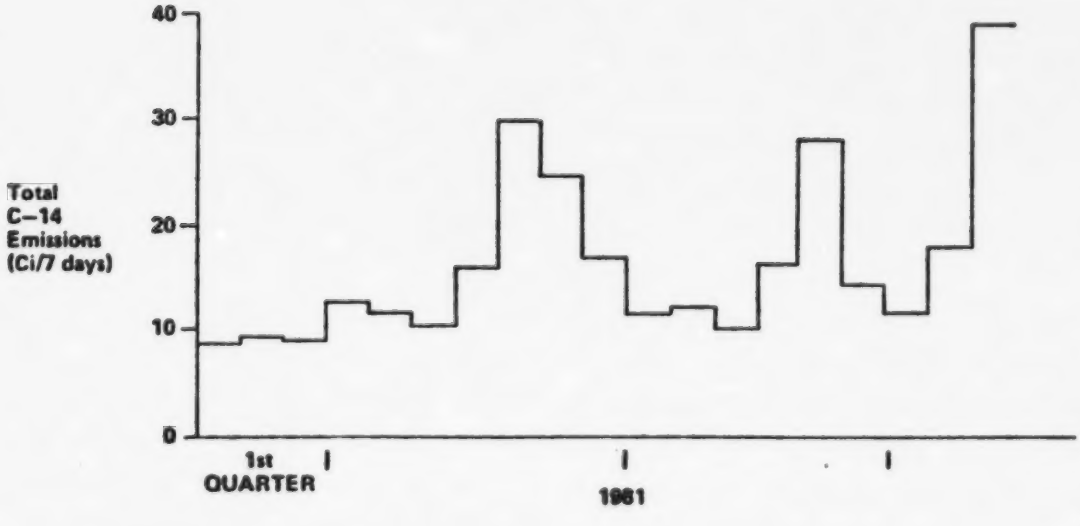

FIGURE 25 TOTAL EMISSIONS OF C-14 AT PNGS 'A' (7)

emissions were observed on three occasions. The magnitude of the peaks indicated that an inventory of C-14, which was larger than predicted by measured system concentrations, was stored somehow in the station. The timing of the emission of these inventories appeared to coincide with major outages. As the normal emission rates from the affected units were less than the production rate in the annulus gas system, it might be suggested that an inventory could be established in the annulus gas system. This observation is not well understood (7).

Figure 26 illustrates various carbon-14 normalized production and emission rates for different types of reactors found globally (7). Comparison indicates that, although production rates are much higher, C-14 emissions from BNGS 'A' type reactors are not much greater than emissions from other types of power reactors. Although emissions from PNGS 'A' are greater (as much as 0.93% of the PDEL) it is expected that these will be substantially reduced by the conversion of the N_2 annulus gas to CO_2. This change is currently being engineered for two units (7).

In general, all radioactive emissions were below 1% of the DEL for the stations. This conforms with the performance targets adopted by the operators of the stations.

2.2 Specific Airborne Emission Points and Pathways

The "open concept" Bruce Nuclear Generating Station 'A' (BNGS 'A') plant is depicted in Figure 27 (8). The four reactor vaults, turbines and generators are all enclosed in a single building shell without any major dividing barriers. The reactors share a common containment envelope and each reactor vault is surrounded by confinement rooms (Figure 27). Most heavy water systems are located within reactor containment or in the confinement rooms, with only a few components located in open areas.

Chronic low level escapes of tritiated heavy water from their containing systems give rise to airborne tritium contamination (8).

The principal component of airborne contamination control is the placement of the majority of heavy water systems within the reactor containment structure. The design, operation and maintenance of this structure ensures that these contained systems are a minimal source of both environmental emissions and occupational exposures. However, some heavy water systems which must be accessible at power are located outside containment, mostly in confinement rooms around each reactor.

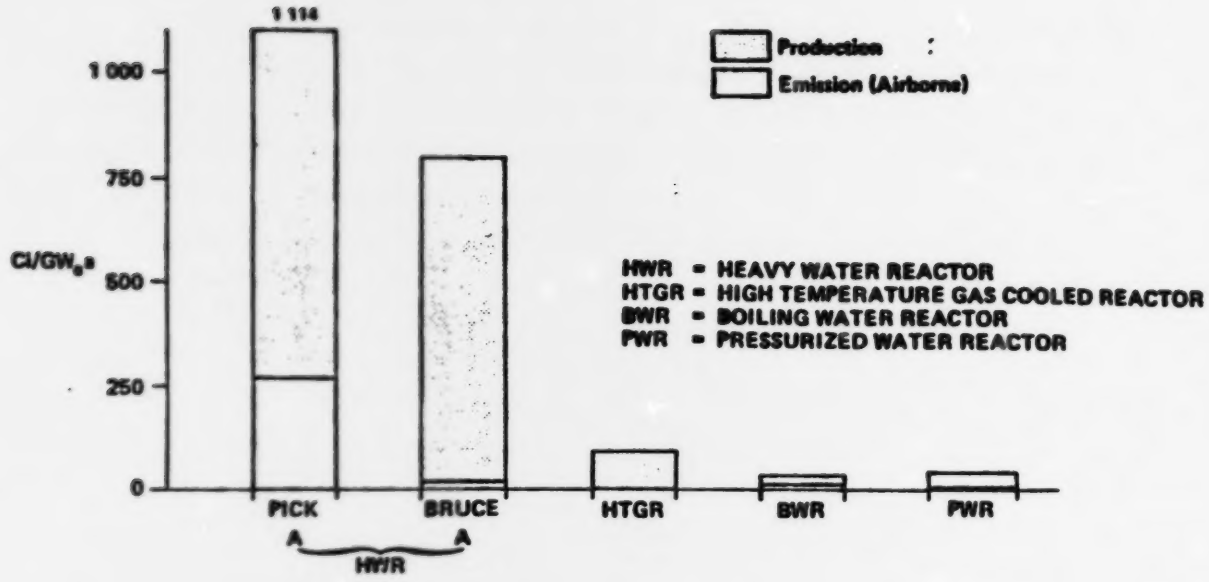

FIGURE 26 COMPARISON OF C-14 PRODUCTION AND EMISSION FOR WORLD REACTOR TYPES (7)

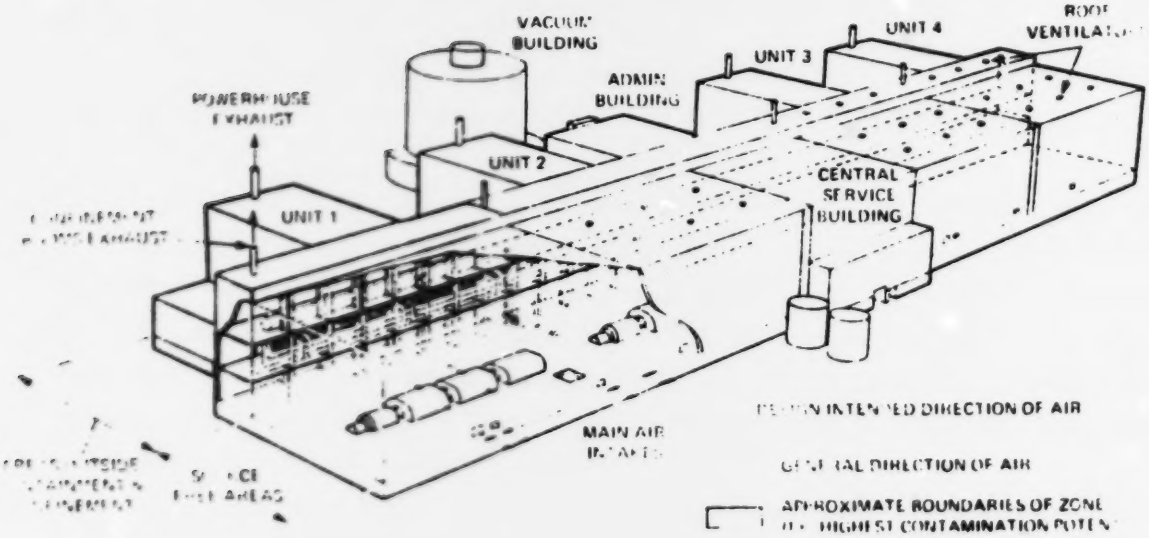

FIGURE 27 SCHEMATIC CROSS-SECTIONAL VIEW OF BRUCE NGS "A" (8)

The reactor containment is fitted with a closed cycle heavy water vapour recovery system which will collect tritium oxide vapour, while confinement rooms are fitted with local purge ventilation only (8). Station air quality (exclusive of containment and confinement) is maintained via purge ventilation designed to move air from the least contaminated areas to the most contaminated areas.

Figure 28 is a schematic which illustrates the basic ventilation layout and the relative magnitudes and directions of air velocities within the plant. Both Figures 27 and 28 indicate the confinement rooms exhaust, powerhouse exhaust and roof ventilators.

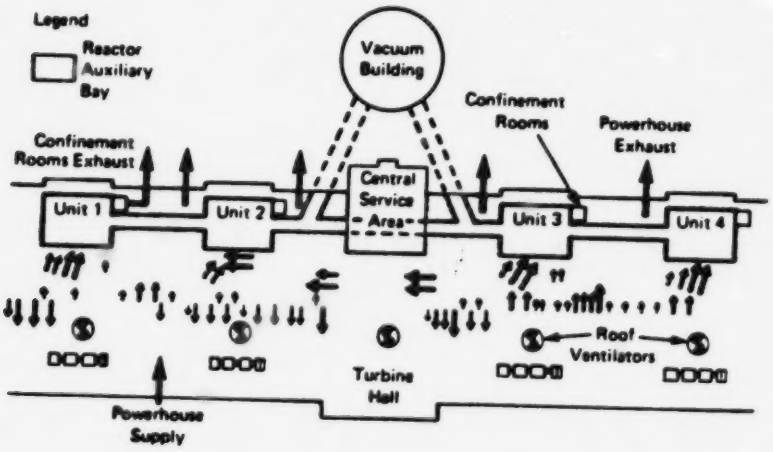

FIGURE 28 BASIC VENTILATION LAYOUT AND AIR VELOCITIES (8)

Figure 29 is a detailed schematic of the gaseous waste management system (9). Zone 3 areas are defined to be the highest radiological areas of the station and include the reactors, irradiated fuel bays and all areas containing "radioactive systems". Zone 2 is a transitional zone where there may be periodic contaimination due to movement of radioactive materials through this area. This includes the Turbine Hall and portions of the Central Service Wing. Zone 1 is considered to be totally non-active and is accessible to non-atomic radiation workers.

Air exhaust from the Zone 3 areas is handled in either one or two systems; the "contaminated" or "noncontaminated" exhaust system. For the Bruce and Darlington type stations the contaminated exhaust-system for each unit typically exhausts up to 43 m^3/sec of air, via duct work, from areas known to contain sources of airborne activity.

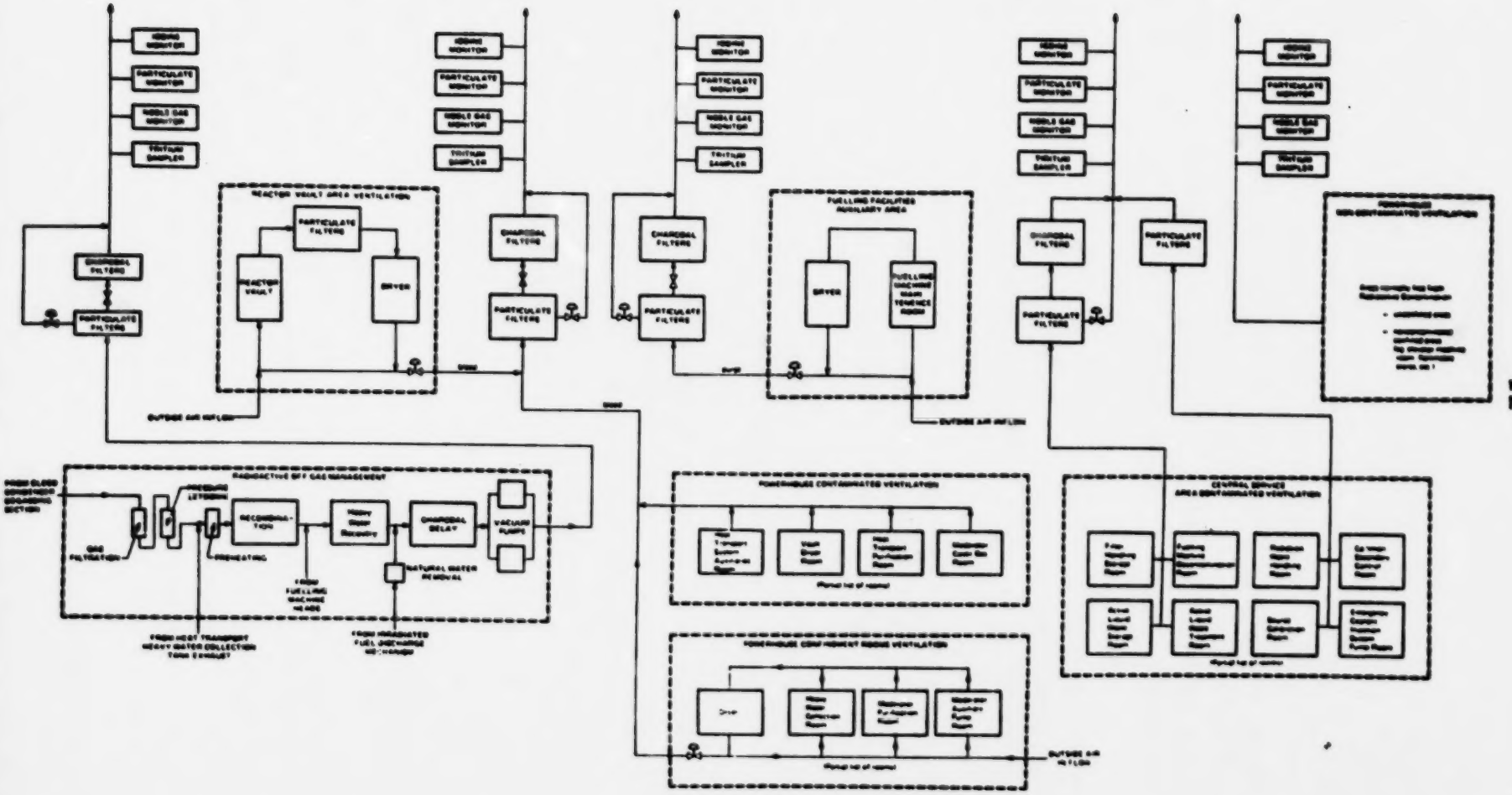

FIGURE 29 RADIOACTIVE GASEOUS WASTE MANAGEMENT (9)

This includes the many confinement rooms located adjacent to the reactor building, containing auxiliary heavy water systems. Also included is the relatively small purge flow for pressure balance from the containment exhaust drier system. Air flow into the confinement rooms is controlled via the exhaust system to minimize back flow and spread of airborne contamination to other parts of the station thereby maintaining good breathing air quality. The system is designed to assist air movement from less contaminated to more contaminated areas.

The exhaust from the contaminated ventilation system passes through a filtration unit consisting of a prefilter, HEPA filter and a TEDA impregnated charcoal bed (described in detail in section 4) prior to stack monitoring for particulates, iodine-131, noble gases and tritium.

Separatorless HEPA filters are used so that they can be volume reduced prior to storage as solid radwaste. HEPA filters are DOP* tested twice per year and the removal efficiency of the charcoal filters is checked four times per year via the use of test cannisters.

In addition, there is a non contaminated exhaust system from the Zone 3 area of up to 240 m^3/sec that is monitored but not filtered prior to release. This is exhaust air from the reactor auxiliary bay areas where little contamination is found and there is at present no requirement to filter to achieve operating emission targets. The air to be exhausted from the Zone 2 area (which includes the Turbine Hall) is periodically sampled to ensure that there is no significant activity emission via this pathway.

Another source of radioactive air emissions is the Bruce Incinerator. All of the routine reactor operating wastes from Pickering NGS 'A' (and eventually Pickering NGS "B" and Darlington NGS "A") are transported through mainly rural areas to the Bruce Nuclear Power Development for processing and storage (9).

At the Radioactive Waste Operations Site, the combustible and compactible wastes are processed at the Waste Volume Reduction Facility (WVRF) and the non-processible waste is sent directly to storage.

Since 1977, the combustible waste category has been volume reduced, prior to storage, in a batch pyrolysis type starved air incinerator. The system consists of a primary chamber, to which 15 m^3 (2000 kg) of waste is top charged per batch, and where the waste is pyrolyzed at a temperature of 500°C. Combustion is completed in a propane fired afterburner chamber where volatile gases and particulate carry over are burnt off at a temperature of 900°C before they enter the flue gas treatment system. The flue gas is

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*DOP or, more correctly, DEHP (di-2 ethylhexyl phthalate) is a filter-test aerosol.

cooled to 200°C by dilution with air and by an air to air heat exchanger. The flue gas is then passed through a baghouse before it is released to the stack.

At present waste up to 0.2 mSv/h (20 mrem/h) is incinerated but the vast majority has less than 0.05 mSv/5 (5 mR/h) at 30 cm. Ash from the primary chamber is unloaded by gravity discharge into 2.5 m^3 rectangular metal containers immediately after the end of the burn cycle. An average batch-load of waste produces about 0.4 m^3 of ash, giving an average gross volume reduction factor of 40:1. Specific activity of the ash collected, ranges from 18×10^7 to 370×10^7 Bq/m^3 (5 to 100 mCi/m^3). The net stored volume reduction factor is about 25-30:1. Contact fields on most of the ash containers is between 0.1 to 0.2 mSv/h (10 to 20 mrem/h). Emissions from the stack have been low, averaging 2.2×10^4 Bq (0.6 μCi) of gamma particulate activity released per m^3 of waste burned.

The radwaste incinerator system is a working prototype, and as such, has had problems due to some basic design deficiencies. Nevertheless, the operation has been relatively productive, accumulating over 20 000 operating hours to date and having processed over 8000 m^3 waste, of which 2800 m^3 was burned in 1980. The process, control and equipment have been steadily modified during the course of operation. One of the main deficiencies has been the excessive length of the burn cycle (40 hrs. compared to design 24 hrs.) which limits the maximum incineration capacity to about 3000 m^3 of waste per year. Other problems encountered were: flue gas heat exchanger corrosion and plugging (somewhat alleviated by upstream partial air dilution cooling); failure of automatic control mode of operation; excessive propane consumption; and deformation of primary chamber inner steel liner (a portion of which has been replaced with a refractory lining).

Ontario Hydro is currently studying installation of a new radwaste incinerator by 1985. More incinerator capacity will be required to be able to process all of the projected future combustible waste arisings. A continuous cycle type, proven, radwaste incinerator is favoured.

2.3 Specific radionuclides

(a) Tritium

During operation within the reactor core, neutron activation converts some of the deuterium in heavy water to the radioactive isotope of hydrogen, tritium. At present (Sept. '83) the average tritium concentrations in the moderator and heat transport liquids at the Bruce 'A' generating station are about 15 Ci/kg and 1.5 Ci/kg respectively (10). In

the absence of tritum removal measures, it is anticipated that after 30 years of operation the moderator concentration level could be about 50 Ci/kg and that of the heat transport system about 2 Ci/kg (10).

A field study was carried out at Bruce NGS 'A' (8). A random selection of the inventory of potentially leaking components was field surveyed for heavy water leakage using a vapour phase Infra-red (IR) Absorption Spectrometer tuned to a D_2O absorption line. The IR absorption spectrometer is capable of detecting leaks of 10 mg/day. Approximately 150 of the 5000 process system components of one reactor unit were leak tested. It was found that practically all nonwelded mechanical joints, flanges, valves, and level gauges used in heavy water service leaked to some extent, although all leakages measured were well below the visual detection level of 20 g of D_2O/hour (Table 4). The leakage rate distribution appeared skewed, since most of the leaks detected were small, and only a few components showed major leaks. Of the components studied, 70% were found to leak less than 0.1 g/day. Those components which leaked in excess of 1.0 g/day contributed to 95% of the total amount of D_2O leakage measured. In Table 4 the contributions of the various component classes to the total station chronic D_2O leakage are shown. It can be seen that tube and pipe fittings (Compression Joints, etc.) which represent over 60% of the population, contribute less than 6% to the total leakage, while stem sealed (gate) valves, which make up only 0.3% of the components, contribute almost 36% of the leakage.

To study the transport of airborne tritium, mappings of station air movement patterns were carried out, using a propeller type anemometer capable of detecting air velocities down to 0.1 m/s. The consequences of component leakage and contamination transport were addressed by measurements of tritium-in-air concentrations, using molecular sieve traps (Figure 30). A measurement of tritium-in-air concentration consisted of a week long continuous collection of moisture in the air at selected locations and the subsequent analysis of the collected moisture to determine its tritium concentration. A volume of approximately 1 m^3 was sampled in each measurement and three such measurements were performed for each of 50 locations. The use of these samplers in conjunction with standard liquid scintillation counting techniques permits detection of 10^{-11} Ci/m^3 of tritium (10^{-5}(MPC)a).*

*(MPC)a means Maximum Permissible Concentration in Air. The public (MPC)a for tritium is 300 nCi m^{-3} (300 000 pCi m^{-3})

TABLE 4 RESULTS OF HEAVY WATER LEAK RATE MEASUREMENTS (8)

Components	Total Inventory (%)	% of Total Station Chronic D_2O Leakage	Selected For Testing (No)	Leak Rate <0.1 g/day (%)	Leak Rate >0.1 g/day (%)	Cold Systems - Average Leakage (g/day)
Tube & Pipe Fittings	62.2	5.4	45	91.1	8.9	0.014
- Compression Joints	35.5					
- Plug End Cap	3.3					
- Diaphragm	0.1					
- Quick Connect/Disconnect	0.2					
- Threaded Connections and Other	23.1					
Instrumentation & Control Components (I&C)	4.1	1.4	19	85.7	14.3	0.052
- Temp. Transmitters	1.3					
- Flow Transmitters	1.3					
- Pressure Transmiters	0.9					
- Other	0.6					
Flanges	5.4	28.1	20	55	45.0	0.810
- $\leq$ 4" Dia.	2.9					
- $>$ 4" Dia.	2.5					
Valves	26.9	59.8	57	78.7	10.7	N/A
- Bellow Sealed (BS)	9.1					0.2
- RV (relief)	0.2					0.2
- Gate & Ball with Stem Packing	6.3	(35.9)			(10.6%> 1 g/day)	16.8
- Diaphragm	2.5					0.111
- Instrument Valve with Steam packing and Compression Tube Joints	7.1					0.22
- NRV (Check)	0.4					0.2
- Instrument Valve "Zeroleakage" with Bellows, Butt Welded	7.3				0.003	
Slight Glasses	0.4	0.8	7	57.1	42.9	0.3/meter of length
- Level Gauges	0.2					N/A
- Flow Gauges	0.2					N/A
Septa	0.02	4.5	2	0	(50%> 1 g/day)	9.13
Other - (Strainers, Pump Assemblies Tanks & Pressure Vessels, etc)	0.98			No data available		

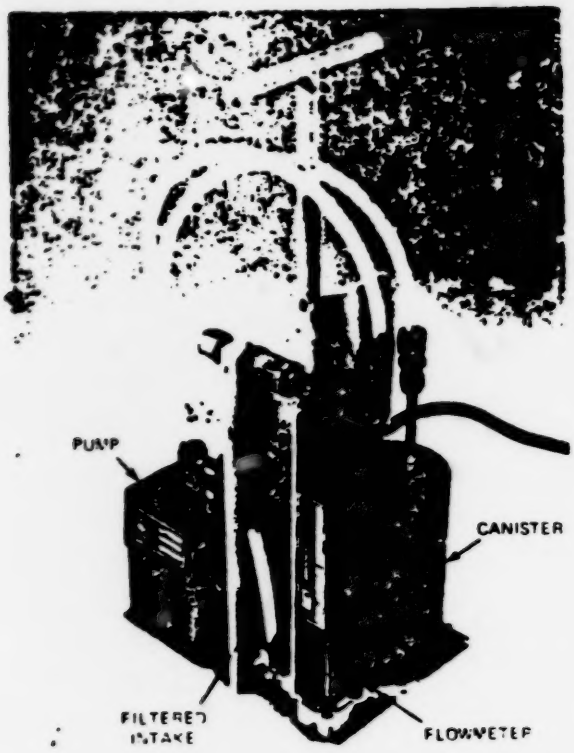

FIGURE 30 MOLECULAR SIEVE SAMPLER USED IN BRUCE NGS 'A' TRITIUM CONTROL FIELD STUDY (8)

Figure 31 illustrates the distribution of tritium levels in the three major functional areas of the plant. Some tritium was detected in all areas surveyed and the concentrations correlated with the proximity of heavy water containing equipment.

Although the average concentrations of tritium-in-air did not exceed the design targets* indicated in Figure 31, local concentrations did exceed these targets in some areas. As expected, the highest tritium concentrations were typically near areas with moderator related equipment. In addition, future contamination concentrations are expected to increased in rough proportion to the tritium activity increases in both heat transport and moderator liquid heavy water.

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*These targets were derived by considering the exhaust flow rates and station occupancy factors (8).

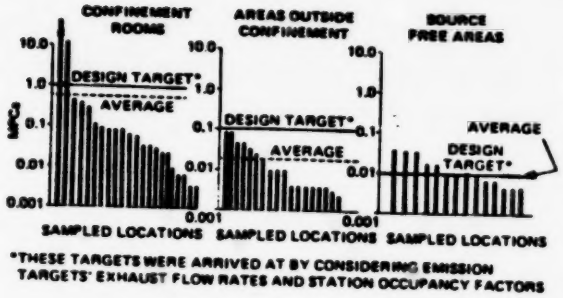

FIGURE 31 AVERAGE TRITIUM CONCENTRATIONS (MPC)a - TEST RESULTS (8)

(b) Radioiodines

Table 5 lists the main fission product iodine isotopes (11). The radioiodine of primary concern in CANDU reactors is iodine-131. It is found as any of several compounds such as molecular iodine, I_2, methyliodide, CH_3 and hypoiodous acid, HOI (12).

TABLE 5 MAIN-FISSION PRODUCT IODINE ISOTOPES (11)

Name	Half-Life	Fission Yield	Main Radiations Emitted (MeV)	
I-129	1.59×10^7 yr	0.9%	β 0.15	γ
I-131	8.04 day	2.77	β 0.6	γ 0.36
I-132	2.28 hr	4.13	β 0.8	γ 0.67
I-133	20.8 hr	6.77	β 1.27	γ 0.53
I-134	52.6 min	7.19	β 2.42	γ 0.85
I-135	6.58 hr	6.7	β 1.0	γ 1.28

Radioiodines are released from defective fuel bundles into three operational systems:

- primary coolant heavy water
- irradiated fuel discharge mechanism
- irradiated fuel inspection and storage bays water

The transfer of radioiodine from leaking primary coolant heavy water to airborne forms is reported as insignificant and the volatility of iodine from the irradiated fuel bay water is efficiently controlled with hydrazine (13). However, significant amounts of airborne radioiodine, released from defective fuel into the Irradiated Fuel Transfer Room atmosphere, migrate to operational areas and subsequently to effluent streams.

(c) Noble Gases

The radioactive noble gases may be fission products or activation products. The fission products of importance are isotopes of krypton and xenon. The activation product argon-41 may be produced from neutron activation of argon-40 normally found in air.

Noble gases released to the environment are an external radiation hazard. The most important noble gases from an environmental perspective are presented in Table 6 (14).

TABLE 6 IMPORTANT NOBLE GAS RADIOISOTOPES (14)

Radionuclide	Half Life	($\bar{E}_\gamma$ (Mev))	($\bar{E}_\beta$ (Mev))
^{41}Ar	1.83 h	1.29	0.46
^{85}Kr	4.4 h	0.179	0.238
^{85}Kr	10.76 y	0.00283	0.233
^{88}Kr (^{88}Rb)	3.8 h (17.8 min)	2.822	2.383
^{133}Xe	5.27 d	0.03	0.11
^{135}Xe	9.14 h	0.267	0.313
^{138}Xe (^{138}Cs)	17.5 min (32.2 min)	2.81	1.85

The production rate of argon-41 is determined by station design. At Pickering NGS 'A' where nitrogen rather than air is used as the annulus gas between calandria and pressure tubes, the production rate is much lower than that at Douglas Point NGS (15). The most abundant species in the gaseous effluent are usually xenon-133, xenon-135 and possibly argon-41. Krypton-88 and caesium-138 emit highly energetic radiations but are very short lived. The long lived krypton-85 has very low fission yield and very low mean gamma energy ($\bar{E}\gamma$). It may have to be considered only from the point of view of long term accumulation in the atmosphere on a global scale.

Radioactive noble gases are normally contained in the fuel and heat transport system. They escape to building atmospheres in the same ways as radioiodines. This results in airborne contamination in some areas. The very tight heat transport system and closed cycle drying systems with small exhaust flows effectively delay noble gases prior to release to the environment.

A central noble gas off-gas treatment system is now installed at each major station with the object of reducing emissions associated with fuel failures (see section 5.1) (9).

(d) Particulates

These fission products may escape from the fuel bundles in the reactor, due to imperfect cladding, or in the irradiated fuel bay. However, fuel sheath failures or defects are few in number and increasingly uncommon. It is more difficult for fission products to escape in particulate form due to fuel sheath defects than as gaseous fission products. A number of fission product noble gases decay to particulate material. Gaseous fission products may also escape by molecular permeation through an intact fuel sheath.

The radionuclides of greatest concern in this category are listed in Table 7(14).

TABLE 7 IMPORTANT PARTICULATES (14)

Radionuclide	MPCa (Ci/m^3)	
	Inhalation	Food Chain (milk)
Cs - 137	1.5×10^{-9}	5×10^{-11}
Cs - 134	1×10^{-9}	1.5×10^{11}
Sr - 90	4×10^{-11}	1.5×10^{-12}
Sr - 89	2×10^{-10}	1.5×10^{-11}
Co - 60	9×10^{-11}	NA
Ru - 106	4×10^{-11}	NA

(e) Carbon-14

Carbon-14 is produced in all water-cooled nuclear powered reactors during normal operation. It is mainly produced by the ^{17}O (n,α) ^{14}C reaction with oxygen in the coolant. Other reactions include ^{14}N (n,p) ^{14}C with nitrogen-14 present in the fuel as

impurities or in the channel annulus as a filling gas, direct activation of carbon-13, ^{13}C (n,γ) ^{14}C and ternary fissions.

Due to the unique design of CANDU reactors, the production of carbon-14 in the moderator and coolant is up to 100 times that of comparable light water reactors (16). Reactions of the carbon-14 atom with surrounding molecules eventually produce hydrocarbons, carbon dioxide and carbonates. Carbon-14 in the form of carbonate (CO_3^{--}) or bicarbonate (HCO_3^-) may be adsorbed onto ion-exchange resin for possible treatment.

The concern about carbon-14 is due to its extremely long half-life (5730a) and its potential for incorporation into the food chain.

The production rate of ^{14}C in CANDU reactors has been estimated to be 0.42 Ci (1.6×10^{10} Bq) per MWe.a, 95% being produced in the moderator (17). However, a more recent estimate showed that the production rate in various pressure tube reactors is between 0.53 and 0.93 Ci (2×10^{10} and 3.4×10^{10} Bq) per MWe.a, depending on the annulus gas used. The presence of a gaseous annulus between the pressure and calandria tubes can substantially increase the overall production of ^{14}C in the core of CANDU reactors. A nitrogen-filled annulus may increase the overall production of ^{14}C by a factor of approximately two, with respect to reactors with carbon dioxide annulus (17). Consequently, more use of CO_2 as an annulus gas is being made and the production of ^{14}C from the annulus becomes a small fraction of the total production in the reactor.

The place of origin of ^{14}C within the reactor has a strong influence on the discharge pathway. Three locations of ^{14}C generation can be identified:

(a) the reactor coolant and moderator where the major fraction is produced
(b) the fuel
(c) structural and other materials of the core.

The chemical form of carbon has been determined at Pickering NGS 'A' by several grab samples (7). Carbon was found to be in the form of carbonates in moderator D_2O (~99%) and CO_2 in the moderator cover gas (>95%). In the nitrogen annulus gas system, however, the chemical form varied: approximately 80% CO_2 and 20% CO in units 3 and 4 and approximately 20% CO_2, 20% CO, and 50% CH_4 in Units 1 and 2 (7).

Most emissions would be expected to travel from the source into containment areas, recirculating vapour recovery systems and then via a purge stream to the reactor building exhaust and the stack.

Carbon-14 produced in annulus gas is rapidly lost to the environment due to system purging or leakage. At Nuclear Power Demonstration Nuclear Generating Station

(NPD NGS) and Douglas Point Nuclear Generation Station (DPNGS), emissions result from purging of the air which forms a combined annulus gas and reactor vault atmosphere. The small volume of the system determines that a dynamic equilibrium would be established quickly and only a small inventory would be retained in the stations (7).

Carbon-14 produced in moderator water can be either removed by the ion exchange resin of the purification system or transferred to the cover gas. Carbon-14 transferred to the cover gas will eventually be lost to the environment due to periodic purging of this system.

Carbon-14 production rates calculated for two operating CANDU reactors, 540 MWe units at Pickering NGS and 750 MWe units at Bruce NGS are listed in Table 8. The CANDU production rates are calculated from core-averaged Westcott fluxes and 200 m/s cross sections of 0-235 b, 1-81b and 0.9 b for the reactions described at the beginning of this section (18).

TABLE 8 ESTIMATED PRODUCTION RATES OF ^{14}C IN VARIOUS REACTOR TYPES (18)

	Carbon-14 Production Rates - Ci/GW(e)a			
Reactor	Coolant	Other Systems - Ci/GW(e)a		Fuel (Average Values)
CANDU - 540 MW(e)	9	Moderator D_2O	440	15
		N_2 Annulus System	430	
750 MW(e)	10	Moderator D_2O	547	20
		CO_2 Annulus System	0.02	
BWR	4.7	Cladding & Struct. Mater.	43.3-60.4	17.6
PWR	5.0	Cladding & Struct. Mater.	30.5-41.6	18.8
LMFBR	nil	Cladding & Struct. Mater.	12.8	6.3
HTGR	nil	Cladding & Struct. Mater.	<190	12

3 MONITORING OF AIRBORNE EFFLUENTS

3.1 General

Stack effluent monitoring at Ontario Hydro's nuclear power stations presently consists of particulate and radioiodine collectors with on-line control monitors and a noble gas monitor in series. Tritium sampling is separately provided in parallel with the above system. The system is relatively complex, its design has not been optimized for either compliance or control monitoring, and operational experience indicates that improvements are needed. Control, compliance and emergency monitoring objectives and programmes have been defined, a new concept of stack effluent monitoring systems has been developed, and effluent monitoring performance standards are being established (19).

Figure 32 illustrates a gaseous monitoring system (19). It is based on an optimized sampling line with particulate and iodine collectors that are continuously monitored during the sampling period and then measured in a laboratory for compliance evaluation. The system consists of:

(a) A holder containing a glass fibre filter particulate collector and an impregnated charcoal cartridge radioiodine collector connected in series.

(b) A shielded scintillation detector (NaI) sensing both collectors.

(c) A gross gamma monitor with the lower level discriminator set at approximately 320 keV to eliminate ^{133}Xe + ^{135}Xe, and reduce external gamma background interference.

(d) A push-button-controlled single-channel analyser set at the ^{131}I energy (364 keV $\pm$ 10%).

(e) Two monitoring functions:

(i) count-rate from the collected radionuclides;

(ii) increment of the collected activity, evaluated in short time periods (the increments are directly proportional to radionuclide concentration in the effluent stream and hence their emission rate).

(f) Monitor alarms preset for:

(i) total activity collected (from count-rate meter);

(ii) emission rate (from time-controlled differential scaler).

3.2 Tritium

Tritium monitoring is complicated because of the very low energy beta particles emitted (E max = 18 keV). Continuous tritium-in-air monitors generally consist

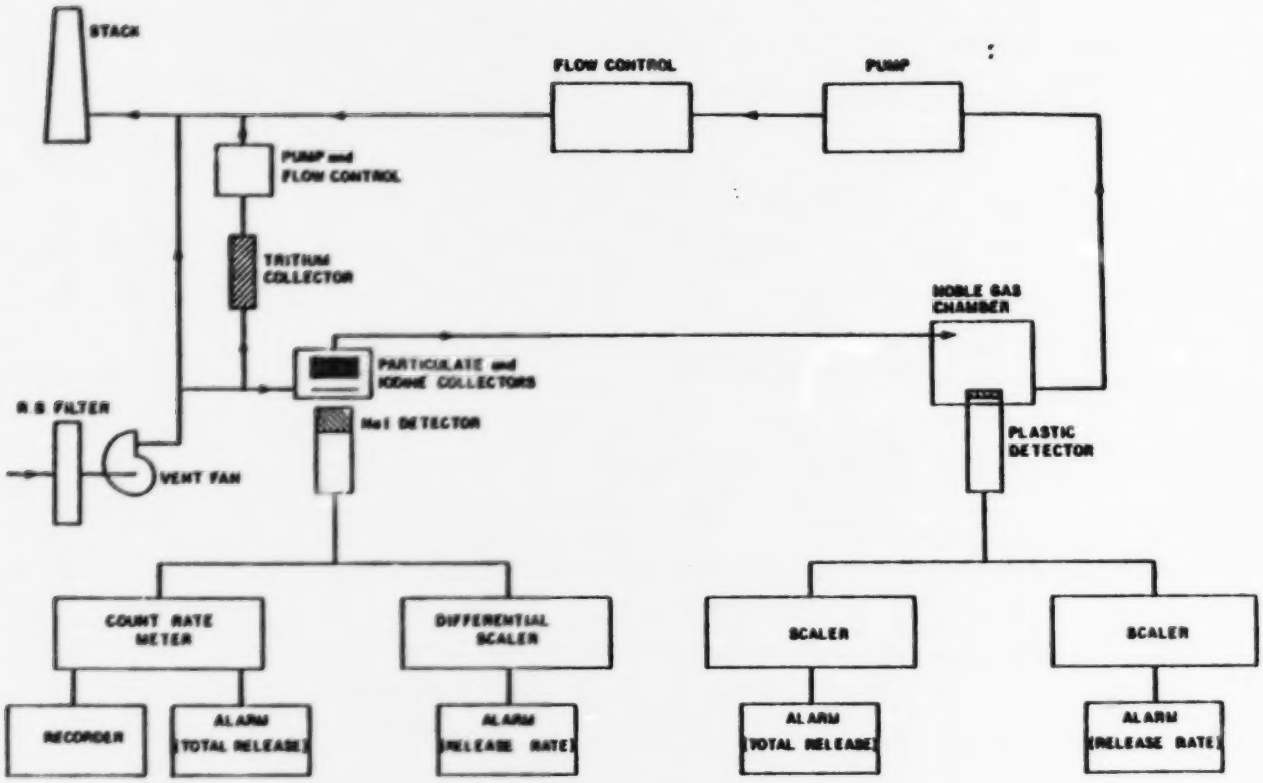

FIGURE 32 NEW CONCEPT OF GASEOUS EFFLUENT MONITORING (19)

of gamma compensated ionization chambers (Appendix B). Such instruments despite compensation, are subject to interference from local gamma sources and noble gases and from experience have been found impractical for stack monitoring. The best method for tritium oxide has been to extract a sample from the gaseous effluent and to remove the tritium from the sampled stream. Two methods have proven successful (14):

(i) Bubblers

A sample from the effluent gas is continuously bubbled through 250 mL of demineralized water using a standard gas washing bottle and a small diaphragm pump. Flow is controlled using an adjustable flow rator and the sampling period is 24 hours. A 1 mL sample of the bubbler water is then counted using a liquid scintillation counter. The tritium released is determined according to the following:

$$\text{Tritium released per 24 hours (Ci)} = \text{Tritium Collected in Sampler per 24 h (Ci)} \times \frac{\text{Exhaust duct flow rate}}{\text{Sampling flow rate}}$$

(ii) Silica Gel Samplers

A sample of effluent gas is passed through a sealed glass tube containing silica gel for a 24 hour sampling period. Care is taken to adjust the flow rate to a level that will not saturate the gel. A 1 mL sample is then counted in the scintillation counter to determine the tritium oxide released. The sampler is usually weighed before and after use to determine the average moisture content of the gaseous exhaust, an important measurement in heavy water management.

The silica gel samplers are preferred for compliance monitoring for the following reasons:

(a) The silica gel sampler is prepared as a shelf item and no operator action is needed to prepare it.

(b) There is no evaporative loss of sampler contents as is the case with bubblers used for long term sampling in high temperature areas.

(c) There is no problem with loose lids allowing in-leakage of air from other than the sample stream.

The silica gel samplers have the disadvantage that they require a longer analysis time than bubblers.

Results of snow and tree sap sampling for tritium near the Bruce Nuclear Power Development (BNPD) are presented in Table 9. Additional measurements of tritium activity in cell-free foliage extracts are given in Table 10 and isopleths are shown in Figure 33. In all cases declining tritium concentrations were observed with increasing distance from BNPD (20).

3.3 Radioiodines

3.3.1 Chemical Forms. The compositon of airborne iodine in operational areas and effluents has been evaluated using a species selective sampler (21). Hypoiodous acid was the major airborne species, measured in air samples through several weeks after discharging defective fuel elements from a reactor. The relative content of organic forms increased with time. Typical ratios of iodine species, measured in Ontario Hydro nuclear power plants are given in Table 11 (22).

TABLE 9 TRITIUM, 1982, NEAR THE BNPD, DOUGLAS POINT (20)

Sampling Site Number	Tritium Concentration in Melted Snow (n Ci/L)	Tritium Concentration in Tree Sap (n Ci/L)
5	20	9.7
7	5.0	7.0
8	6.9	7.0
9	30	6.5
10	6.2	4.3
11	3.0	2.5
12	4.0	-
13 (S)	3.3	5.4
13 (N)	3.9	
14	1.4	2.3
15	9.6	-
16	2.7	-
17 (Pt. Elgin)	1.8	-
18	9.7	-
19	14	12.7
20	7.8	-

Note Canadian Target for Drinking Water is 108 nCi/L.
U.S. EPA limit for Drinking Water is 20 nCi/L
(based on 4 mrem from 2 L daily ingestion).

TABLE 10 TRITIUM ACTIVITY IN CELL-FREE FOLIAGE EXTRACTS COLLECTED IN THE VICINITY OF THE BNPD, DOUGLAS POINT, ONTARIO, SEPTEMBER 1981 (20)

Sampling Site	Tritium Activity (measured as activity in nCi, per litre of water extracted)	Measurement Error (±)
2	17.5	0.2
3	4.12	0.13
4	10.9	0.2
5	8.99	0.17
6	2.53	0.12
7	2.12	0.12
8	1.70	0.11
9	2.52	0.12
10	1.37	0.11
11	1.41	0.11
12	0.96	0.26
13	2.94	0.12
14	0.97	0.11

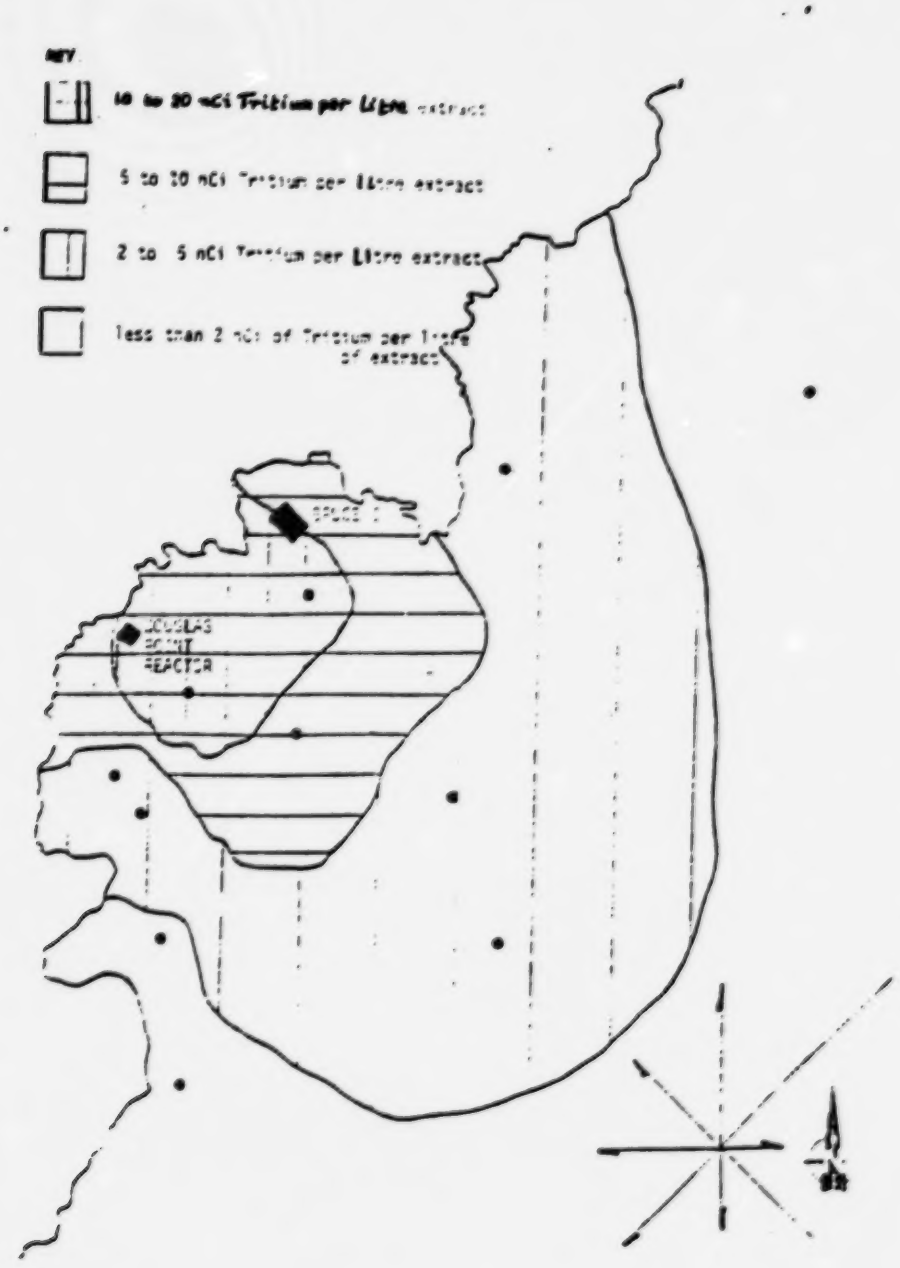
KEY
10 to 20 nCi Tritium per Litre extract
5 to 10 nCi Tritium per Litre extract
2 to 5 nCi Tritium per Litre extract
less than 2 nCi of Tritium per litre of extract
DOUGLAS POINT REACTOR

FIGURE 33 **ISOPLETHS SHOWING TRITIUM ACTIVITY IN CELL FREE EXTRACTS IN THE VICINITY OF BNPD, DOUGLAS POINT (20)**

TABLE 11 AVERAGE COMPOSITION OF AIRBORNE RADIOIODINES IN CANDU POWER PLANT AREAS (22)

Airborne ^{131}I Concentration (MPC)a (occup.)	Particulate I %	I_2 %	HOI %	Org. I %
25 - 300	~ 15	1 - 5	55 - 75	10 - 20
~ 2	~ 10	~ 5	50 - 65	20 - 30
~ 0.1	~ 10	~ 5	30 - 40	40 - 60

It is assumed that the ratio of airborne iodine species under LOCA conditions in CANDU reactor containment will be similar to the above ratio, measured in the range of 25-300 Maximum Permissible Concentration in Air (MPC)a.

No measurable Cs-137 was found in airborne samples, which contained high levels of radioiodines. This indicates that short lived radioiodines, including ^{131}I, are released from irradiated CANDU fuel as elemental iodine and possibly some HOI.

3.3.2 Field Performance of Radioiodine Adsorbents

For defined operational conditions and performance requirements, the optimal types of adsorbent were recommended for Normal Operation Assemblies (NOA) and the air cleaning assembly for emergency and cleanup operation (ECA) respectively. Since tryethylenediamine (TEDA) impregnated (or coimpregnated) charcoal has a superior performance under room temperature and high humidity conditions, and also its aging is slower than the aging of KI_X charcoal, it has been exclusively used in the NOAs, which are in continuous operation in the Ontario Hydro power plants. However KI_X impregnated charcoal is required in the stand-by ECAs, because of its high ignition point and negligible desorption of impregnant at elevated temperatures.

The performance standards that have been applied at Ontario Hydro since 1976 based on the results of experimental evaluation of iodine deposition on grass (22) are shown in Table 12. The NOA adsorbers have been tested in 6 month intervals at Pickering NGS for both CH_3I and HOI efficiency. Since sample canisters were not available with the NOAs installed before 1979, charcoal has to be sampled at the outlet face of the bed with a "slotted-tube sampler", according to the ASTM E300 standard. Despite relatively large sampling errors, the evaluation of large numbers of test results enabled a determination of the relative humidity effect on the rate of impregnated charcoals aging in adsorber units, exposed to continuous air flow. The results are summarized in Table 13.

TABLE 12 THE MINIMUM REQUIREMENTS FOR IODINE REMOVAL EFFICIENCY. (derived from the results of experimental evaluation of iodine deposition on grass) (22)

Stage	Contaminant	Minimum Design Value Decontamination Factor (DF)	Minimum Efficiency - %	Iodine Deposition Rate - $m.s^{-1}$
HEPA	0.3 µm DOP	1250	99.97	--
Adsorber	I_2 vapour	1000	99.95	8×10^{-3}
	HOI	670	99.90	7×10^{-4}
	CH_3I	33	97.00	1×10^{-5}

Test conditions: 25°C, 95% RH, air velocity 20 cm/sec, adsorbent depth - 50 mm.

The aging trends generally confirm that TEDA impregnated charcoal is more suitable for use in the NOA. Operational lifetimes of charcoal in specific air cleaning assemblies have been estimated from the data in Table 13, to optimize the procurement of replacement charcoal.

TABLE 13 CHARCOAL AGING IN CONTINUOUS AIR FLOW (22)

Ventilated Area	Average RH %	Average Temp °C	Charcoal Impregr.	Average rate of penetration increase %/25mm/a CH_3I	Average rate of penetration increase %/25mm/a HOI
Reactor Containment	20	20	5% KIx	13	--
	20	20	5% TEDA	8 - 10	0.25
Irrad. Fuel Bay	90	20	5% KIx	25	1.0
	90	20	5% TEDA	15	0.25

3.4 Noble Gases

3.4.1 Gamma Ci-MeV Monitoring of Noble Gas Effluents. It can be shown that the external gamma dose rate from a cloud of noble gas is essentially proportional to the mean gamma energy per disintegration of the radionuclide under consideration i.e. it is proportional to the gamma - Ci - MeV (15). Measurement of gamma - Ci - MeV is there-

fore the parameter required in a gaseous effluent containing a noble gas mixture if boundary gamma dose rate is the relevant criterion. At Pickering NGS the derived emission limit (DEL) for noble gas radioiosotopes is 4.3×10^4 gamma-Ci-MeV per 7 day period,* based on a maximum permissible concentration in air ((MPC)a) for whole body exposure of 6.4×10^{-8} gamma Ci-MeV per cubic metre (14).

The gamma-Ci-MeV values for the most common noble gas isotopes in nuclear power stations effluents are listed in Table 14 (19).

TABLE 14 GAMMA Ci-MeV VALUES OF NOBLE GAS RADIOISOTOPES IN GASEOUS EFFLUENTS (19)

Radionuclide	Decay half-life	$\bar{E}_\gamma$(Ci-MeV/disint.)
^{41}Ar	108 min	1.29
$^{85}Kr^m$	4.4 h	0.156
^{87}Kr	76 min	0.775
$^{88}Kr + ^{88}Rb$	2.8 h	2.03 + 0.692
^{133}Xe	5.27 d	0.030
$^{133}Xe^m$	2.26 d	0.023
^{135}Xe	9.2 h	0.245
$^{135}Xe^m$	15.6 min	0.421
$^{138}Xe + ^{138}Cs$	17 min	1.053 + 2.387

The gamma C_i - MeV content of the noble gas in the effluent can be monitored in two ways:

(a) Using an energy corrected gamma radiation detector with detection efficiency directly proportional to energy of detected gamma radiation. The pulse rate from this detector is proportional to the 'gamma C_i - MeV concentration' of the noble gas mixture in the detection chamber and integrated pulses are proportional to the total noble gas gamma C_i - MeV emission.

(b) Using a scintillation detector which has an output pulse proportional to the detected gamma energy. The detection current is then directly proportional to the gamma C_i - MeV concentration of noble gases in the detection chamber.

*This value has now been revised to 4.5×10^{14} gamma Ci-MeV per 7 day period (6).

Both of these methods require that only the gamma component of the noble gas radiation is monitored and that constant effluent flow is maintained throughout the monitoring period.

3.4.2 Gamma Energy Response of a GM Detector (19). The gamma energy response of the Amperex type 912 NB3 GM detector presently used in noble gas monitoring may be measured using a set of National Bureau of Standards (NBS) primary gamma standards covering the energy range of 60 keV - 1.3 MeV (^{241}Am, ^{109}Cd, ^{57}Co, ^{203}Hg, ^{133}Ba, ^{137}Cs, ^{54}Mn, ^{60}Co). The energy response is measured with the lead shielding originally supplied on the GM detector, and with the shielding removed. The energy response correcting effect of several materials is tested in order to optimize the detector for noble gas Ci-MeV monitoring. The optimum energy response is achieved using a combination of 6 mm plexiglas, 2 mm of Cu and 1 mm of steel shielding around the GM tube.

The noble gas detector and flow chamber assembly are calibrated using gaseous ^{41}Ar and ^{133}Xe samples.

Figure 34 shows the Ci-MeV response curves measured using the previously listed NBS sources. It also indicates that the counts per Ci-MeV for ^{133}Xe and ^{41}Ar, two major radionuclides in CANDU power station effluents, with the above composite shielding are almost identical. In the gamma energy region of approximately 0.12 MeV the detector Ci -MeV response is 50% greater than that measured at ^{133}Xe and ^{41}Ar energies.

3.4.3 Application of Thermoluminescent Dosimeters (TLDs) for Noble Gas Emergency Monitoring (19). The noble gas monitors used presently in CANDU stations have the following disadvantages:

(a) Limited upper monitoring range (long GM-detector dead time);
(b) Unsatisfactory energy response for Ci-Mev measurement;
(c) No accumulative sample is available for accurate laboratory evaluation of noble gas emissions;
(d) Information on released noble gas radioisotopes may be lost from power loss or accidental resetting of the scaler.

The applicability of TLDs for noble gas monitoring has been investigated. Their gamma energy response characteristics and wide detection range indicated that they could be employed as a back-up to the on-line stack effluent noble gas monitors, particularly for emergency purposes.

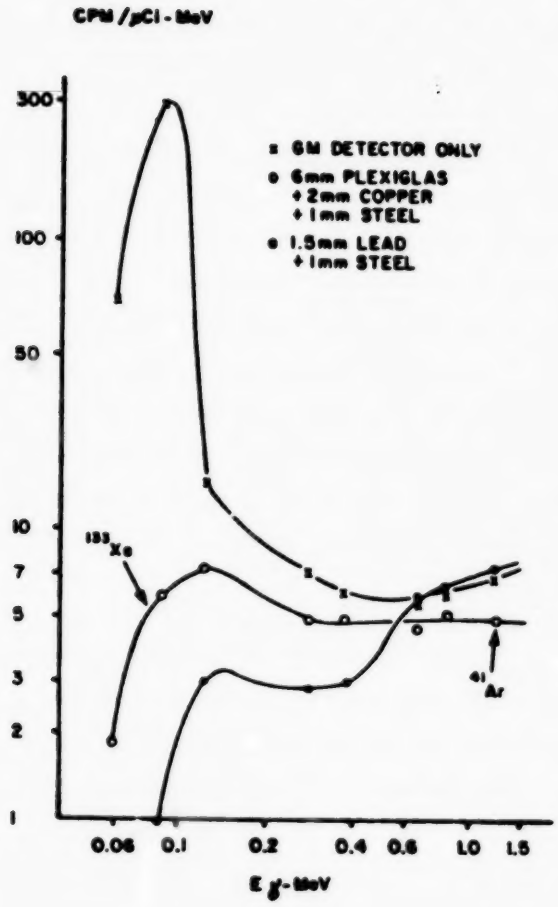

FIGURE 34 GAMMA ENERGY RESPONSE OF THE AMPEREX TYPE 912 NB3 GM DETECTOR (19)

An experimental set-up has been installed in the Pickering NGS ventilation stack. Both the noble gas gamma radiation in the stack and external gamma background were detected coincidentally with two sets of TLDs and periodically evaluated. The noble gas effluent was coincidentally measured with a gamma spectrometer and the C_i - MeV totalled for each TLD evaluation period. It was concluded that TLDs in such an arrangement can measure noble gas emission levels of <1% of emission limits with good accuracy.

3.5 Particulates

Gross particulate activity is measured by collection of the particulate activity from a sample of the effluent on an absolute filter which is continuously measured by geiger or scintillation detector and count rate meter. At Pickering, instrument sensitivity is such that a sample flow of 56 litres/minute enables a gross particulate emission of 2.5 μCi/day to be measured reliably in an effluent gas flow of 185 m^3/minute (12).

At Gentilly (12), the quantification of the emission is performed in the laboratory by measuring the absolute filter with a geiger detector. Instrument sensitivity is such that a sample flow of 112 litres/minute enables a gross particulate emission of 0.5 μCi/day to be measured reliably in an effluent gas flow of 1780 m^3/minute.

Since it is impractical to measure all individual radionuclides, the particulates are monitored as a mixture with their emission limit set to the most restrictive component, caesium -137* (5) and the gross beta-gamma activity is measured.

The derived emission limit (DEL) in this category has not changed from 1 Ci/7 days at Pickering NGS 'A' despite the change in standard from $^{90}Sr + ^{90}Y$ to ^{137}Cs.

3.6 Carbon-14

3.6.1 Reactor Systems. Carbon-14 has been identified in reactor systems as a mixture of CO_2, CO and hydrocarbons (18). Their ratio depends on the chemistry of the respective reactor system. The predominant mechanism appears to be oxidation of carbon to CO_2 and CO by oxygen radicals formed from radiolytic decomposition of the coolant water.

An annulus gas sample of nitrogen from a CANDU reactor was analyzed in the NY State Department of Health Laboratory. In this sample more than 75% of carbon-14 was present as hydrocarbons, as expected, because of the presence of D_2 in the annulus gas (from the coolant). Samples of primary coolant off-gas (D_2) and moderator cover gas (He) have not yet been analyzed but from chemical conditions it may be assumed that hydrocarbons are the major carbon-14 compounds in the primary coolant and significant portions of carbon-14 in the moderator system are in oxide forms.

Carbon-14 produced in UO_2 fuel might become bound to uranium as carbide, remain as impurity atoms, or be converted to carbon monoxide or dioxide.

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*Formerly, the emission limit was based on the assumption that all released activity is due to a combination of ^{90}Sr and ^{90}Y (19).

3.6.2 Present Status in Monitoring and Control of Gaseous Carbon-14 Emissions. The practice of CO_2 purification with heated CaO, applied in some carbon dating procedures, has been further developed for direct collection of gaseous carbon-14 in nuclear facilities. The well known method of CO_2 collection in alkaline solutions has been utilized on several occasions for carbon-14 sampling. However, this method is not selective for CO_2 since other carbon compounds can be partially absorbed and other radionuclides can interfere with carbon-14 measurement. A heated catalyst must be added when "total ^{14}C" is to be collected and evaporation or spills of alkaline solution can introduce significant experimental errors.

A sophisticated laboratory method has been developed in the NY State Department of Health Laboratory for ^{14}C analysis (18). Carbon-14 gaseous compounds are separated with a gas chromatograph and radiometrically evaluated. This method was successfully applied in PWR and BWR effluents and also a CANDU sample evaluation. Unfortunately, the method is too complex for routine ^{14}C monitoring in the field.

In most field applications, gaseous organic compounds and CO are first oxidized to CO_2 by catalytic combustion. Carbon dioxide absorption on CaO and $Ca(OH)_2$ may be evaluated by injecting $^{14}CO_2$ into an air stream containing approximately 300 ppm of nonradioactive CO_2 and passing the air through test columns containing the absorbents. From the measured $^{14}CO_2$ concentration upstream and downstream of the column, the efficiency and capacity of both CaO and $Ca(OH)_2$ for CO_2 absorption can be evaluated within a wide range of experimental conditions. The temperature and gas purging effects on $Ca\text{-}CO_3$ dissociation may then be evaluated by continuous measurement of $^{14}CO_2$ concentrations in the purge gas downstream of the column and also by collecting CO_2 in NaOH solution followed by liquid scintillation counting of ^{14}C.

3.6.3 Field Applications. Based on the experimental work of Kabat (18) measurements of total C-14 emission rates were made by continuously sampling stack gases. The type of field sampler used is shown in Figure 35 and the results are presented in Figures 24 and 25.

Carbon dioxide in the sample gas was collected in the presence of a catalyst on dry calcium hydroxide absorbent at an elevated temperature (950 °C). Hydrocarbons and CO which passed the first collector were catalytically oxidized and collected on a second absorption bed. The collectors were changed every two weeks and the collected C-14 was then extracted separately from each collector into NaOH bubblers and analysed. The error in each measurement was estimated to be aproximately ± 25 % (7).

The advantages of CO_2 collection on solid $Ca(OH)_2$ are:

(i) Selective $^{14}CO_2$ or "total ^{14}C" sampling may be performed.

(ii) The method has good potential for ^{14}C removal (Section 5.2). Large amounts of ^{14}C can be solidified directly during collection. The highly concentrated ^{14}C as $CaCO_3$ can be hermetically sealed for long term disposal.

The main drawback with this type of monitor is that it is difficult to maintain the solid calcium hydroxide absorbent and it is too complex for constant use.

Simpler bubbler systems have been investigated and a prototype (Figure 36) based on sodium hydroxide has been developed and evaluated at PNGS-A over a period of 22 weeks (7). The efficiency of carbon dioxide collection by the first bubbler was $\geq$ 99% with very little CO_2 appearing in the second bubbler. So far this has produced comparable results to those obtained using the calcium hydroxide based experimental monitor.

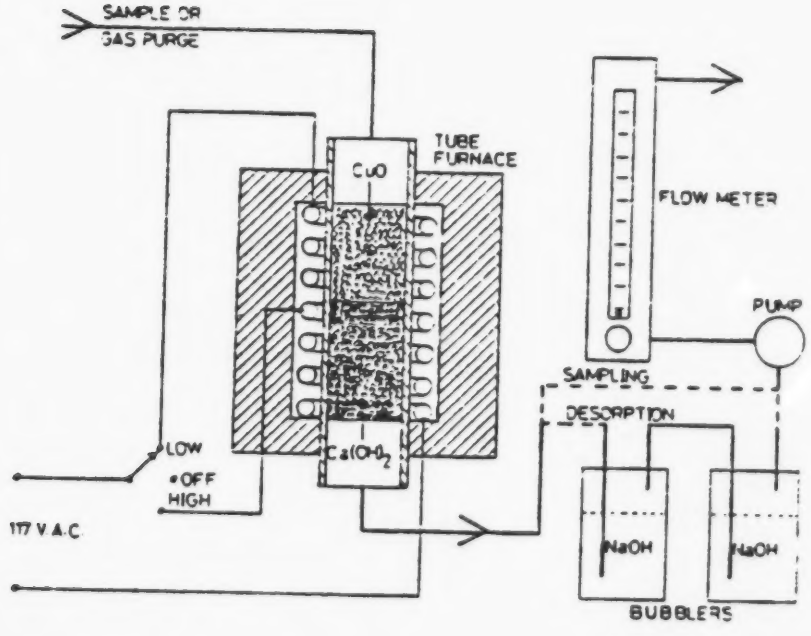

FIGURE 35 EXPERIMENTAL FIELD SAMPLER FOR "TOTAL ^{14}C." (18)

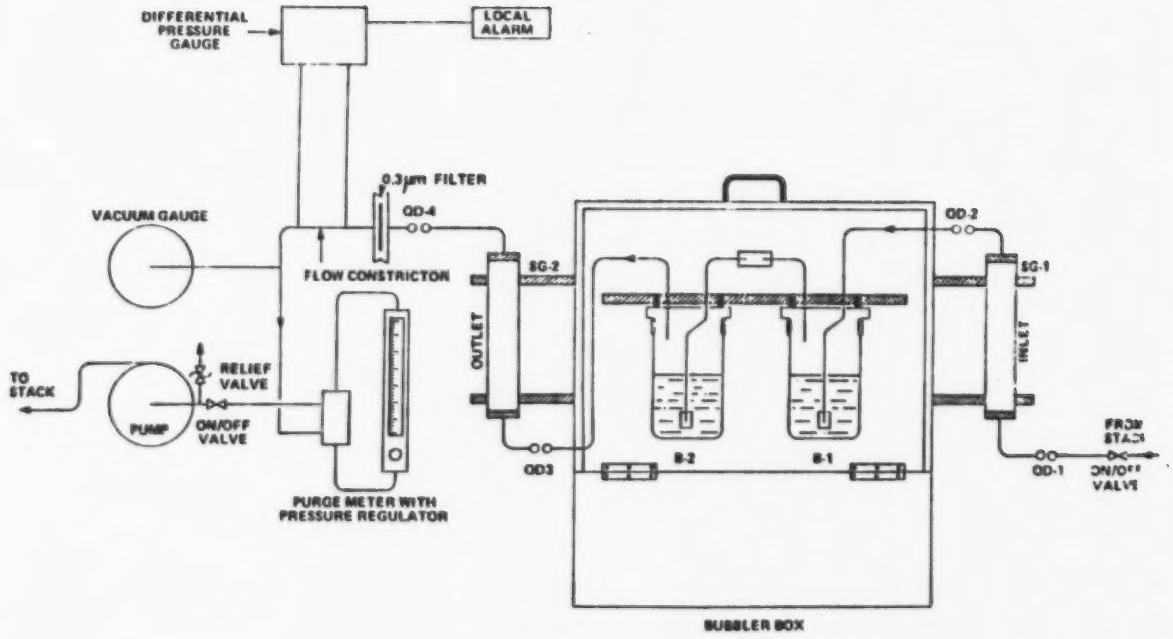

FIGURE 36 SCHEMATIC FOR PROPOSED $^{14}CO_2$ COMPLIANCE MONITOR

3.6.4 Studies of ^{14}C Concentrations in Terrestrial Biota around the Pickering and BNPD Sites (23). A method was developed using oxygen stream combustion, collection of the combustion products and absorption of a known mass of carbon dioxide in an organic absorber. This is followed by liquid scintillation counting for carbon-14.

Samples of terrestrial biota from the environs of Pickering and Bruce were analysed for ^{14}C. Near the Pickering site boundary it was found that ^{14}C concentrations were six times nominal background.

The concentrations of ^{14}C and isopleths for BNPD and Pickering are presented in Figures 37 to 40.

The tree ring analyses at Pickering show a correlation between station capacity factor and above background ^{14}C concentrations in vegetation.

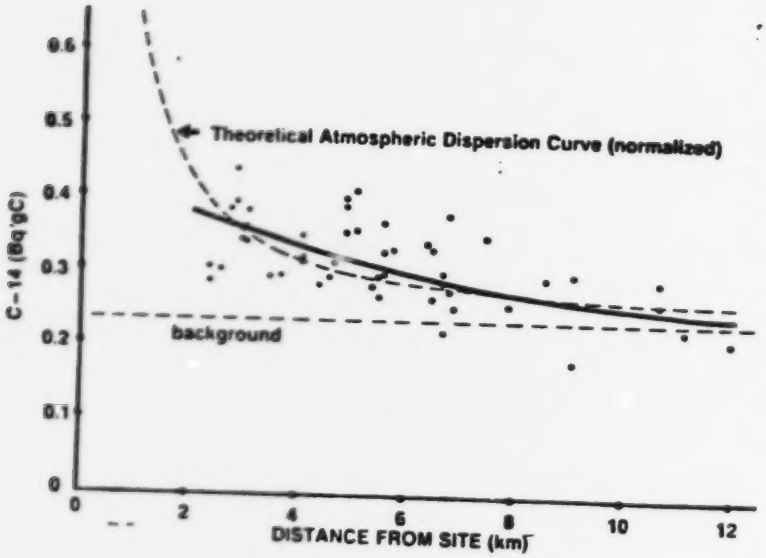

FIGURE 37 C-14 IN BRUCE VEGETATION VS DISTANCE FROM SITE (23)

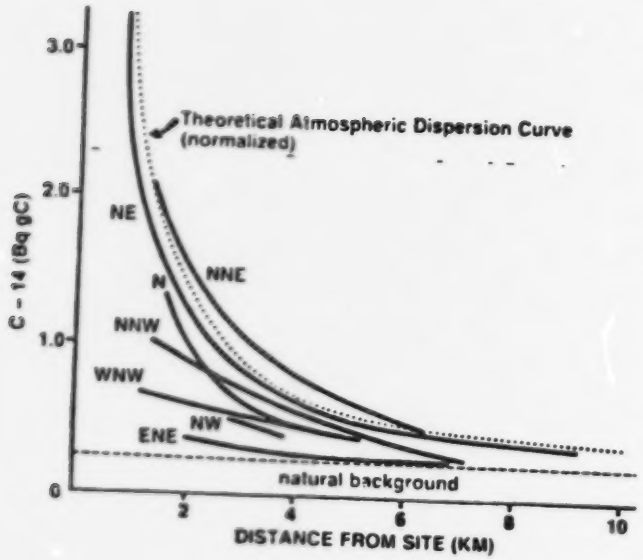

FIGURE 38 C-14 IN PICKERING VEGETATION VERSUS DISTANCE FROM SITE (23)

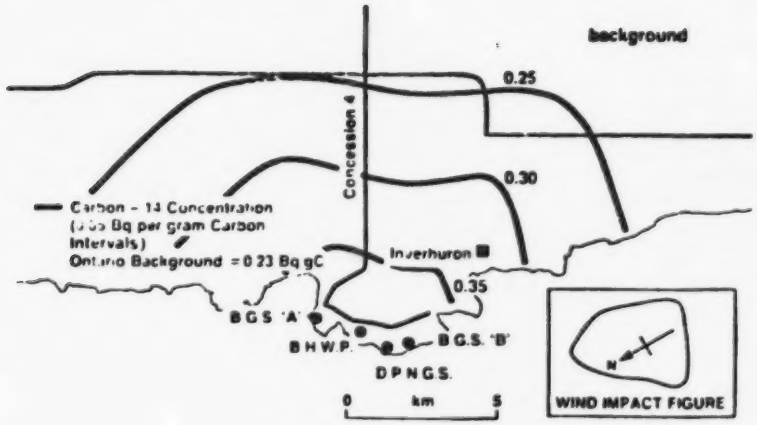

FIGURE 39 CARBON-14 CONCENTRATION IN VEGETATION BNPD AREA ISOPLETHS (1980 - 1982 AVERAGE) (23)

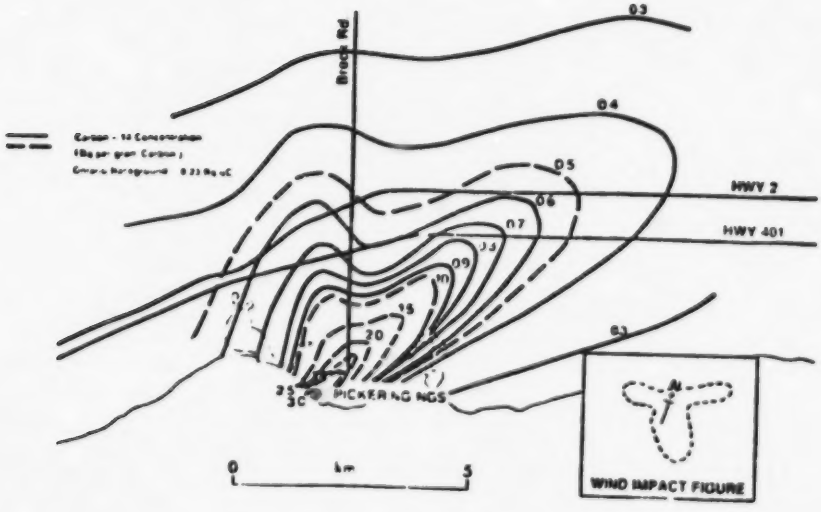

FIGURE 40 CARBON-14 CONCENTRATION IN VEGETATION PICKERING AREA ISOPLETHS (1980-82 AVERAGE)(23)

4 SPECIFIC TECHNOLOGIES FOR CONTROL OF AIRBORNE EMISSIONS

4.1 General

The radionuclide formation and emission characteristics of CANDU power reactors are considerably different from those of light water PWRs. The major difference in emission terms at normal operation results from the on-power refuelling feature of the CANDU reactor (fission products) and its higher rate of formation of neutron activation products, tritium and carbon-14.

To ensure that emissions are as low as reasonably achievable during normal operation, several gaseous control systems have been implemented in CANDU power plants. Gaseous radionuclides are removed in three stages of their migration pathway from their sources to the emission points (22).

The first stage is to remove the radionuclides from operational systems. All dissolved fission and activation products are continuously removed from the primary coolant, moderator heavy water and irradiated fuel bay water with ion exchange purification systems. Noble gas radioisotopes are collected from the primary coolant cover gas and the irradiated fuel discharge mechanism with off-gas systems, filled with activated charcoal.

In the second stage radionuclides are removed from operational areas. As previously discussed (Section 2.3(b)), the release of radioiodine from the Irradiated Fuel Bay water to the atmosphere is efficiently reduced with hydrazine addition during the discharge of defective fuel elements. Tritium oxide vapour is collected by heavy water recovery units from the atmosphere in all areas containing primary coolant and moderator systems. Tritium will also be removed from process heavy water with an isotope separation system.

The third stage is the removal of radionuclides from ventilation streams. At present the removal of noble gas radioisotopes is considered impractical (22). However, other radionuclides are effectively removed from gaseous effluents with high efficiency particulate air (HEPA) filters and iodine adsorbers under both normal and accidental release conditions.

4.2 Containment Building

A description of the containment building has been given in Section 2.2.

4.3 Particulate Removal

The primary component of a nuclear plant exhaust is the High-Efficiency Particulate Air (HEPA) or "absolute" filter. Most systems also include a prefilter, and some reactor and radiochemical plant systems will also include activated charcoal adsorbers for trapping radioactive gases, moisture separators or both.

HEPA filters are by far the most satisfactory and economical devices for removing submicron particles from air and gas streams at extremely high collection efficiency. A very detailed description of the design and performance of such filters is given in ref. 24. A HEPA filter is a throwaway, extended-medium, dry-type filter having:

(1) a minimum particle removal efficiency of no less than 99.97% for 0.3 μ particles
(2) a maximum resistance when clean of 250 N/m^2 (25.4 kg/m^2) when operated at rated air flows capacity
(3) a rigid casing extending the full depth of the medium.

Typical construction of wood-cased and steel-cased open-face HEPA filters is shown in Figure 41 (24). The cone (i.e. filter pack) is made by pleating a continuous web of filter glass or cellulose-asbestos paper (the medium) back and forth over corrugated separators which add strength to the cone and form air passages between the folds. The cone is sealed into a full-depth wood or steel casing (frame), usually with an elastometric adhesive.

The filter papers consist of very fine (submicron) fibres in a matrix of larger (1 to 4 μ) fibres. An organic binder (5% maximum, by weight) is added to hold the fibres during the papermaking process. - resistant papers are made from glass fibres. Combustible papers required for certain special applications are made from cellulose and asbestos fibres. A minimum thickness of 3.8×10^{-4} m (0.015 in) and a minimum basis weight, of 21.8 kg (48 lb) per 280 m^2 (3000 ft^2) are sometimes specified to ensure adequate abrasion resistance. The filter paper is extremely weak and fragile, and the filters must be handled with care to avoid damage. Tensile strength is sharply reduced (about 50%) at temperatures above 204°C (400°F) probably due to burnoff of the binder.

Separator materials commonly used are aluminum foil and asbestos paper. Kraft paper and various plastics are also used in some filters but are not recommended for nuclear applications because of poor heat and fire resistance.

The sealant usually used to seal the cone into the case is a heat and moisture-resistant elastomeric adhesive. One filter type uses a chemically expanded heat-resistant

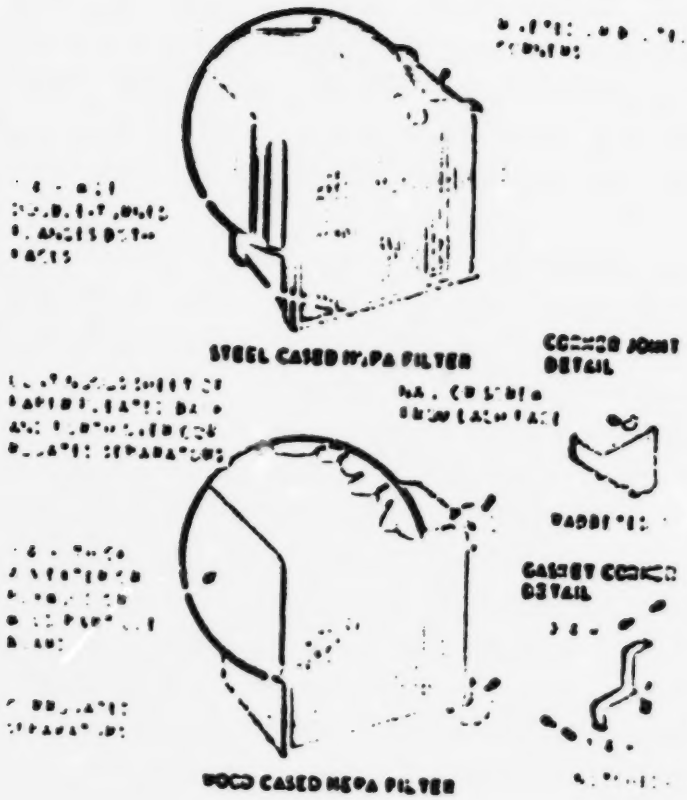

FIGURE 41 CONSTRUCTION OF OPEN-FACE HEPA FILTER UNITS, (24)

urethane foam. Filters that are operated continuously at high temperature (above 204°C (400°F)) are sealed with compressed glass-fibre matting or high-temperature silicone or refractory adhesives: The sealant

(1) must be moisture resistant

(2) must not deteriorate excessively or lose its resiliency under alternating exposure to heat and cold or dry and humid air

(3) must not crack or delaminate from the frame at high temperature (5 min. at up to 399°C (750°F)

(4) must maintain a reliable seal between the filter cone and casing during continuous operation under service conditions.

The high-temperature sealants do not meet all of these criteria, and their use, except in very special circumstances, is not recommended. Glass-fibre matting may not

maintain a reliable seal at high temperature, silicone sealants delaminate quickly at only 10 or 20° above their rated temperatures, and refractory sealants are extremely brittle after heating. Rubber-base adhesives are generally used for gluing the gaskets to the filter. Since the gasket is completely constrained after the filter has been installed, the only requirement on these adhesives is that they hold the gasket firmly until installation of the filter.

The usual casing materials are five-retardant exterior-grade plywood or wood-particle board and cadmium-plated carbon steel. Gaskets are critical items. Tests have shown that excessive variation in gasket-thickness, poorly formed gasket-corners, and improperly glued gaskets result in air leakage exceeding the acceptance level of the filter. When the gasket material is too hard, excessive bolt loading may be required to properly seal the filter to the mounting frame, resulting in possible filter damage. When the gasket material is too soft, excessive compression set may take place, resulting in air leakage as clamping bolts and casing materials relax or expand under service conditions. The best information at this time suggests the use of closed-cell neoprene sponge with cut surfaces on both faces. Gaskets are usually made in strips (1.9 cm x 0.64 cm (3/4 in x 1/4 in)) with notched or dovetailed corners.

Rectangular and cylindrical HEPA filters are available in open-face and enclosed configurations. Open face construction is shown in Figure 41. The enclosed filter (Figure 42) is similar except that the case is longer and closed, with nipples for attachment of the filter as an integral part of the duct. Currently available enclosed filter units do not qualify for fire resistance. Wood-cased enclosed filter units do not meet fire protection standards. Steel-cased enclosed filters, because of their bolted, or riveted corners, may leak under service conditions. Cylindrical filters often provide the solution to some design problems. However, because experience with them has not always been satisfactory and because of fire danger they are not generally recommended for hazardous applications.

Costs

Relative costs (1970 dollars) of various open-face HEPA filter constructions are given in Table 15. Unit costs of 1000 cfm filters (1970) are about $60,000 to $80,000 (U.S.).

FIGURE 42 1000-cfm WOOD-CASED ENCLOSED HEPA FILTER (24)

TABLE 15 RELATIVE COSTS OF VARIOUS HEPA FILTER CONSTRUCTIONS (24)

Casing Material	Separator Material: Untreated Asbestos	Aluminum Foil	Treated Asbestos	Plastic	Stainless Steel
Wood	1.00	1.02	1.08	1.71	7.73
Carbon Steel	1.13	1.16	1.22	1.89	
Stainless Steel	1.96	1.99	2.04	2.69	8.69

4.4 Control of Fission Product Gases

4.4.1 General. Activated-carbon adsorbers, often described as gas filters, are the most satisfactory devices presently available for trapping of fission product gases from nuclear reactors and radiochemical operations. These adsorbers are tightly packed beds of adsorbent carbon granules through which air and gases are passed before release to the atmosphere. The carbon is "activated" by controlled heating under a steam atmosphere, which drives off organic matter and generates large internal surfaces or "sites" on which

adsorption can take place. The internal area of activated carbon varies from 700 to 1800 m^2/g. The carbon is often impregnated with chemicals to increase its affinity for certain gases.

The gas of primary concern in the design of adsorber systems for nuclear applications is elemental radioiodine, which is the most abundant (from the standpoint of radioactivity) sorbable gas released in a reactor accident or in operations with nuclear fuel (24). Under some conditions, organic radioiodine compounds, principally methyl iodide, may also be formed. Although the quantity is small relative to the amount of elemental radioiodine present, it represents a possible health hazard, and it is usually necessary to design the system to remove it. Carbons impregnated with potassium iodide (KI), triethylenediamine (TEDA), or other compounds are necessary for trapping organic radioiodine compounds in humid air.

4.4.2 Performance of Activated Carbon Adsorbers. The important properties of an adsorber are trapping (i.e. adsorption) efficiency, holding capacity or "activity", retentivity or the ability to prevent desorption of the sorbed iodine, ignition temperature, air flow capacity, and resistance to air flow.

(a) **Adsorption Efficiency.** The efficiency of a carbon adsorber is a function of

(1) the degree of activation, ash and moisture content, and impurities in the charcoal
(2) the type and quantity of impregnant (for impregnated carbons)
(3) granule size (the separation factor varies inversely with granule size)
(4) the contact time between the gas and the carbon (i.e. the residence time in the carbon bed, which is a function of bed depth and velocity)
(5) the temperature and humidity of the gas stream.

Special high-purity low-ash grades of carbon are required for the high levels of performance required of nuclear plant exhaust applications. The efficiencies of nuclear-grade charcoals, based on 2.54 cm (1-in.) bed depth for elemental iodine efficiency and 5.1 cm (2-in.) bed depth for methyl iodide efficiency, can be summarized as follows:

(1) Nonimpregnated charcoals: Efficiency for elemental iodine is satisfactory (over 99%) even after extended operation in high-temperature (127° to 138°C (260 to 280°F)) environment containing steam and water droplets. Efficiency for methyl iodide is satisfactory at relative humidities less than 70% but nil at high (over 80%) humidity.

(2) Impregnated charcoals: Efficiency for elemental iodine is satisfactory (over 99%) under all temperature and humidity conditions, up to 132°C (270°F) and 100% relative humidity. Single-pass efficiency with 0.2 sec residence time is given in Table 16. Flooding of the carbon due to condensation or impingement of free water may reduce efficiency to as low as 20%.

TABLE 16 RECOMMENDED DESIGN VALUES FOR SINGLE-PASS METHYL IODIDE EFFICIENCY OF FULL-SCALE ADSORBERS CONTAINING IMPREGNATED ACTIVATED CARBON[a]. 2in. bed depth 0.2 sec residence time, 3 mg radioiodine as methyl iodide per gram of carbon (24)

Relative Humidity (%)	Percent Efficiency for Radioiodine as Methyl Iodide	
	70°F	270°F
85 or less	95	98
90	90	90
95	80	70
98	70	30

[a]R.F. Adams, Oak Ridge National laboratory, personal communication to C.A. Burchsted.

The efficiency of both impregnated and nonimpregnated carbons, for both elemental iodine and methyl iodide, is reduced somewhat when the iodine loading in the gas stream is less than about 0.01 mg/m^3. This report is still under investigation, but preliminary reports indicate that efficiencies are still within the ranges given above.

(b) **Holding Capacity.** When the quantity of iodine charged to a charcoal adsorber exceeds its holding capacity, breakthrough occurs and efficiency drops accordingly. Holding capacity is a function of the amount of charcoal in the system, the number of remaining active sites on which adsorption can take place, and, for impregnated charcoals, the nature, quantity, and condition of the impregnant. The mechanism for capture of elemental iodine is physical condensation (i.e. adsorption) of iodine molecules on the active surfaces of the charcoal. The mechanism for capture of organic radioiodine is apparently a combination of chemical reaction and isotopic exchange, in which the

stable iodine of the impregnant substitutes for the radioactive iodine of the methyl iodide; the nonradioactive methyl iodide goes on through the absorber.

Holding capacity for both iodine and methyl iodide decreases with time because of physical or chemical "poisoning" of the carbon by impurities, particularly by hydrocarbons and water, that occupy active sites on the surface or react with impregnant. The holding capacity of impregnated carbon may also decrease with gradual loss of the impregnant by volatilization. Tests with KI and TEDA-impregnated coke-base carbons indicated a loss of 50% of the initial holding capacity for methyl iodide during 18 months exposure to flowing air, a 50% loss in three years when the beds were exposed to static air (as would be the case in a standby system), and a loss of 50% in five years when the beds were sealed in closed containers. Experience with coconut-shell carbons used in the United States shows a more gradual, but still significant, loss of holding capacity.

The loss in holding capacity for elemental iodine is much slower than for methyl iodide. Beds exposed continuously to flowing air at one installation showed adequate remaining capacity for elemental iodine after four years of service. The loss in holding capacity is not accompanied by a corresponding loss in efficiency provided that the quantity of iodine or methyl iodide charged to the bed is less than the holding capacity of the carbon. The loss in holding capacity indicates

(1) the need for routine analytical tests on samples taken from the beds to determine remaining holding capacity
(2) the need for conservative adsorber design
(3) the necessity of protecting the adsorbers from unnecessary exposure to moisture, hydrocarbons and other poisons.

When poisoning is caused by moisture or short-chain hydrocarbons, some degree of rejuvenation can be accomplished by periodic heating of the adsorbers with warm (90 to 100°F) air. Long-chain hydrocarbons are nearly impossible to drive off in this manner. Exposure to point and solvent fumes can reduce holding capacity to the point that efficiency is reduced in only a few months. The adsorbers must be protected during construction and maintenance operations.

Holding capacity can be severely limited when the carbon is wet, as would be the case in some types of reactors if adequate moisture separators were not provided.

(c) **Retentivity.** Because adsorbers may have to be operated for several days or weeks following a reactor accident, consideration must be given to retention of the trapped gases. The retentivity for a sorbed chemical is usually about 35 to 45% of the

holding capacity. Provided the integrated quantity of iodine or methyl iodide charged to the bed is less than the retentivity limit, desorption is not a significant problem at temperatures below 250 to 300°F. Some iodine loss will take place at higher temperatures, particularly from impregnated carbons, which will also desorb the impregnant and further reduce the capacity for radioactive methyl iodide. The extent of loss is still under investigation. The retentivity factor further emphasizes the need for conservative adsorber design.

(d) **Ignition Temperature.** Adsorption systems must be designed so that the decay heat of collected fission products will not cause ignition of the carbon or overheating to the point that collected fission products (and impregnant of impregnated charcoals) are desorbed. As fission products are deposited in the charcoal, the spot or bulk temperature of the carbon can increase to the ignition point unless adequate air flow or other means of cooling is maintained.

The ignition temperature of new charcoal is a function of the ash content, impurities, internal surface area, granule size and hardness. The lowest ignition of new impregnated coconut-base carbons is about 500°F. The ignition temperature of non-impregnated coconut-base carbons ranges from about 640 to 900°F, depending upon ash and impurity content. With conservative bed design, that is, a bed that contains a large volume of charcoal relative to the air capacity of the system and the amount of iodine to which it can be exposed, the specific loading of fission products will be low, reducing the possibility of spontaneous combustion.

(e) **Air Flow Capacity and Resistance.** Air flow capacity is a function of bed configuration and resistance. Resistance is a function of the size of carbon granules (mesh size), packing density, bed thickness, free area of the granule retaining screens, and air flow velocity. For a given bed design (configuration), resistance varies directly with air flow velocity. Efficiency, at least for methyl iodide, varies inversely with velocity because it is dependent on the contact time between the gas and the charcoal. Operation above the rated capacity of the adsorber is therefore not recommended because of the possible reduction in efficiency, as well as the more obvious penalties of higher pressure drop and operating costs. Assuming no change in air flow velocity and that particulate filters are provided upstream to intercept dust that might otherwise collect on the screens or in the charcoal itself, pressure drop will remain constant over the life of the adsorber.

(f) **Adsorber Design.** The mechanisms involved in trapping radioiodine, particularly methyl iodide, are not fully understood, and research continues. Mass transfer, physical condensation, chemical reaction, and isotopic exchange all take place in varying degrees. Because of these uncertainties and because of the known loss in holding capacity with time, a good deal of conservatism should be exercised in the design of adsorber systems (24).

Following a major accident in a nuclear reactor, the concentration of iodine in the air flowing to the adsorbers may range from as low as 10 ppm to as high as 500 ppm, according to the size of the reactor and the nature of the accident. Of this, from 5 to 10 % may exist as methyl or other organic iodide and the remainder as elemental iodine vapour. Where it can be shown that elemental iodine is the only consideration, the adsorber system should contain enough charcoal so that the maximum amount of iodine that could be charged to the adsorber units (beds) under the worst accident conditions will not exceed 5 mg per gram of charcoal. When methyl iodide must also be considered, the system should contain enough charcoal so that the maximum amount of iodine charged to the units will not exceed 2.5 mg per gram of charcoal, that is, double the amount required for elemental iodine alone. These loading rates are probably much higher than would be experienced in the large adsorber systems used in most power reactors.

To determine the amount of charcoal required for a specific system, the quantity of radioiodine that may exist in the containment following an accident must be estimated. It is generally assumed that one-half of the potential iodine inventory in the fuel can escape to the containment space and that one-half of this amount will plate out on the walls and floor (24). On this basis, 25% of the inventory could theoretically get to the absorbers. Based on these assumptions, the following formula may be used to estimate the amount of charcoal required (24):

$$C = 0.22\ Q \qquad (1)$$

where

C = pounds of charcoal required in the adsorber system,

Q = potential iodine inventory that could be released (grams)

Using this formula, a 1000 MWe power reactor, having a potential iodine inventory of 15 000 g, would require a minimum of 3300 lb of carbon to provide protection against both elemental iodine and methyl iodide. Half of this amount is required it if can be shown that only elemental iodine would be present.

(g) **Construction.** Adsorbers are available in a variety of bed and cartridge designs, of which the bed types are of primary interest for large systems. A bed-type adsorber consists of a layer of carbon granules tightly compressed between perforated metal screens which are enclosed with a metal casing. Figure 43 illustrates a cut away of a typical pleated-bed adsorber and its parts. Since their beds are subject to channeling of the charcoal, a minimum bed thickness of 2.54 cm (1 in) and a maximum velocity of 30.5 cm/s (60 f/m) through the bed (i.e. perpendicular to the screen face) are recommended.

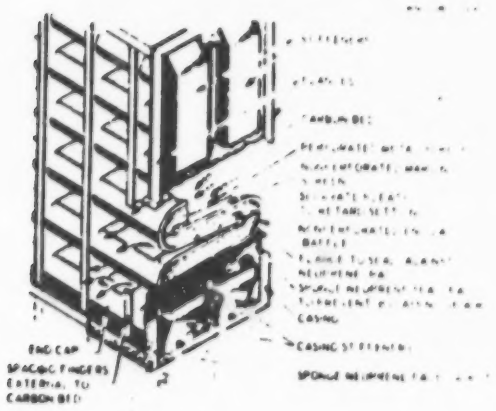

FIGURE 43 CUTAWAY OF 1-in - PLEATED - BED ADSORBER (24)

A single 1-in bed is sufficient when only elemental iodine is under consideration but when, additionally, organic radioiodine (methyl iodide) is present, a bed thickness of 2-in or two 1-in beds in series are required to keep space requirements of the system to a minimum (a residence of the gases in the carbon of ~2 sec. is necessary to develop the required methyl iodide efficiency). The use of two 1-in bed units in series is recommended over a single 2-in. bed unit for two reasons:

(a) the upstream unit will take most of the moisture, impurity and iodine loading, leaving the downstream unit relatively clean (which may reduce total system maintenance costs).

(b) should a leak develop in one unit (resulting from settling of the charcoal, faulty gaskets, cracked welds, etc), the second provides full protection against elemental iodine and a substantial measure of protection against organic iodides.

The cost of two 1-in. bed units does not greatly exceed that of a single 2-in bed unit. Installation costs, however, are substantially higher since two mounting frames and considerably more housing space are required.

5 RECOVERY AND TREATMENT SYSTEMS FOR RADIOACTIVE AIR EMISSIONS

5.1 General

In order to further reduce tritium gaseous emissions below current performance, BNGS'A', BNGS'B' and Darlington NGS'A' (DNGS 'A') stations are having drier systems fitted to some confinement rooms located outside the reactor building containment. Previous practice has been only to dry rooms within containment. It is expected that this will reduce station emissions of tritium in addition to lowering operator doses associated with work in the affected rooms. The installation of tritium removal systems, possibly in the late 1980s, will aid in controlling tritium airborne emissions (9).

A central noble gas off-gas treatment system is installed at each major station with the objective of reducing emissions associated with fuel failures. The system uses ambient temperature charcoal delay beds in series with a charcoal guard bed to prevent poisoning of the main beds. Approximately 35 tonnes of charcoal are used and will never need to be replaced throughout the 40 year lifetime of the system (9). System flow is 22 L/s for the total station and it provides a delay of approximately 34 days before release to the environment. This results in a decontamination factor of 90 for Xenon-133 which is the design basis for the system (9).

The system installed at Pickering NGS 'A' has not been commissioned since fuel, performance and hence noble gas emissions have not necessitated the use of the system. The system at Bruce NGS 'A' has been in operation for more than 4 years and has significantly reduced noble gas emissions. Recent improved fuel performance at Bruce NGS 'A' may reduce the need for continuous use of the system at that station.

Carbon-14, which is predominantly produced in the moderator system, is emitted via the gaseous effluent route. Research and development is currently underway both at Ontario Hydro and AECL to develop methods for removing this gas from moderator water or cover gas systems before it gets on the moderator purification resins or escapes by the cover gas system to station exhaust. This includes the use of calcium hydroxide, barium hydroxide or bismuth oxide resulting in a product that can be immobilized and stored. If the decision is made to implement a removal system, prototype application would be one or two years away (9).

5.2 Carbon-14 Removal System (18)

The proposed method to be used in carbon-14 removal at CANDU nuclear plants is identical to that described in the carbon-14 sampler (section 3.6.3). The material

choice is not critical in this application and so the whole system can be made of carbon steel since its operational temperature does not exceed 450°C and retention of ^{14}C on system components does not have any negative effect.

The choice of catalyst depends on the character of both the carrier gas and carbon-14 compounds to be removed. Commercially available catalyst which can be regenerated is preferable for systems removing ^{14}C components from oxygen free gas.

Only a heated cartridge with an adequate quantity of $Ca(OH)_2$ is required when $^{14}CO_2$ is to be removed from systems or gaseous effluents at a nuclear facility. The recommended loads and temperatures of the absorbent are given in Table 17.

TABLE 17 RECOMMENDED LOADS AND TEMPERATURES OF ABSORBENT (18)

CO_2 Concentration (ppm)	Absorbent Temperature (°C)	Absorbent Load (kg $Ca(OH)_2$ Per kg CO_2 Removed)
> 100	450	3.5
1 - 100	400	4.5
< 1	370	5.0

The recommended purification flow limit is 10 L/S (~20 cfm) per kilogram of $Ca(OH)_2$. This value is independent of CO_2 concentration and gives $\geq$99% efficiency for CO_2 removal throughout the operational period as calculated from Table 17.

The relatively simple removal process does not form by-products or involve handling loose contamination. Carbon dioxide can be removed from high pressure gaseous systems, its adsorption is very selective and thus the used absorbent cartridges will cause negligible gamma fields. No handling of loose Ca $^{14}CO_3$ is necessary, the absorber cartridges can be disposed directly or fixed in concrete for disposal. The high capacity of $Ca(OH)_2$ for CO_2 results in low volume waste to be disposed. The remaining $Ca(OH)_2$ in the disposed cartridge will absorb CO_2, eventually penetrating into the container, and reduce the possibility of ^{14}C leakage.

Other removal processes involving thermal stripping and acid stripping of carbon-14 from an actual resin sample, although unsuccessful are discussed in Ref. 16.

5.3 Noble Gas Recovery and Treatment Systems in Light Water Reactors (25)

5.3.1 General. This section presents options to the system described in section 5.1. In LWR operations, nearly all the noble gas emissions occur in the main condenser steam jet air ejector in boiling-water reactors (BWRs) and from the various primary venting and letdown streams in pressurized water reactors (PWRs). In most LWRs, the noble gases are not separated from the off-gas streams but are delayed and allowed to decay. The more recent designs provide 45 to 60 day holdup systems so that only ^{85}Kr, which has a 10.8 year half-life, remains in any significant concentration after this time.

5.3.2 Waste Gas Treatment Systems in BWRs

System Description.

A typical radioactive waste gas treatment system for the most recent BWR designs is shown in Figure 44. The noncondensable waste gases are drawn from the main condensers with steam jet air ejectors, preheated and passed through hydrogen recombiners to recombine the radiolytically formed hydrogen and oxygen, which constitutes about 80% or more of the waste gas streams. Because of the high exothermic reaction of hydrogen and oxygen, which will result in a temperature rise of ~30°C for every percent of hydrogen in the feed stream, a large recirculation loop is maintained around the recombiner.

After the off-gas from the recombiner is passed through the condenser and excess water is removed, the remaining gas stream is <20% of its volume when it was removed from the main condenser. This gas flow is usually directed through a 30-min delay line to allow the shorter-lived isotopes to decay. The waste gas stream is cooled further and the remaining moisture removed with desiccant dryers. The waste gas in then passed through several large tanks of activated - charcoal adsorbent. Many of the more recent designs include subambient charcoal-adsorption systems to reduce the size of the adsorbent beds.

System Performance

A typical performance of a BWR radioactive waste gas removal system of recent design is shown in Table 18. Over 98% of the total annual emission from a BWR is from the main condenser off-gas. For those BWR's where "clean" steam is supplied to the turbine gland seals rather than to the primary steam, which contains volatile fission products, the percentage of annual radioactive emission via the main condenser off-gas is

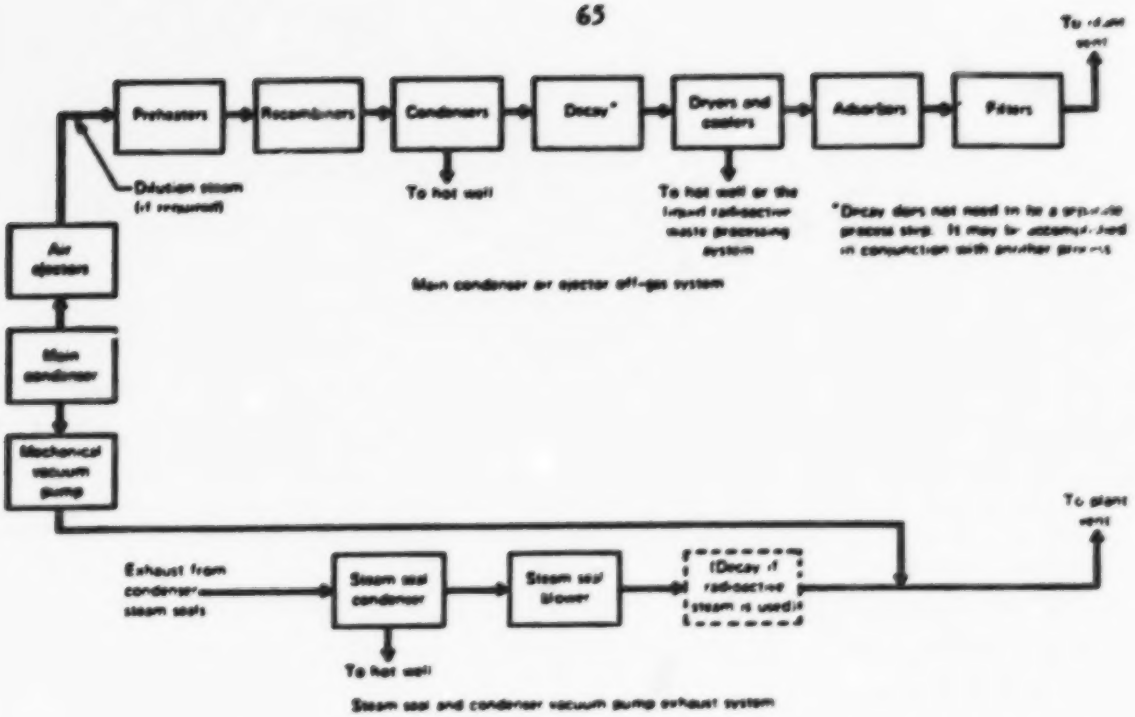
Preheaters
Recombiners
Condensers
Decay*
Dryers and coolers
Adsorbers
Filters
To plant vent
Dilution steam (if required)
To hot well
To hot well or the liquid radioactive waste processing system
*Decay does not need to be a separate process step. It may be accomplished in conjunction with another process
Air ejectors
Main condenser
Mechanical vacuum pump
Main condenser air ejector off-gas system
To plant vent
Exhaust from condenser steam seals
Steam seal condenser
Steam seal blower
(Decay if radioactive steam is used)
To hot well
Steam seal and condenser vacuum pump exhaust system

FIGURE 44 TYPICAL GASEOUS RADIOACTIVE WASTE PROCESSING SYSTEMS IN BWRs (25)

greater than 99%. With a 21 800 kg, activated charcoal, subambient-cooled (-18°C) system, the radioactive waste gas effluent can be reduced to approximately 282 Ci/a, approximately 240 Ci of which is ^{85}Kr.

The estimated emissions from other sources are also shown in Table 18. Any further reduction in these emissions would be very costly and not very cost effective.

5.3.3 Waste Gas Treatment Systems in PWRs (25)

System Description

There are many more different types of radioactive waste gas treatment systems in PWRs than there are in BWRs, but only some of the more common ones are described here. In PWRs, essentially all the noble gases are retained in the primary coolant system and are released only when this system is vented or leaks occur. A much greater inventory of the radioactive noble gases is retained in the coolant system in PWRs than in BWRs. The amount of noble gas emission from PWRs depends on the letdown technique and rate and the venting techniques and rates.

TABLE 18 EFFECTIVENESS OF WASTE GAS TREATMENT SYSTEMS FOR NOBLE GASES IN 1000-MW(e) BWRs (25)

Source of release	Annual release, Ci
Waste gas treatment systems	
With 30-min delay	1 260 000*
With recombiner with subambient charcoal-adsorption delay system† (80-d delay for xenon; 2.7-d delay for krypton)	
Estimated from other sources	282
Containment building	125
Auxiliary building	373
Turbine building	3 637
Radwaste building	1 144
Gland seal exhaust	5 652
Mechanical vacuum pump	1 800

* Of this amount, 240 Ci is ^{85}Kr.
† Based on a 21 800-kg (24-ton) system operating at -18°C (0°F), -29°C (-20°F) dew point, and 36 m^3/h (21 ft^3/min) air inleakage.

Generally PWR waste off-gases are compressed in storage tanks to allow for decay and are then either vented to the atmosphere or recycled. A typical system is shown in Figure 45. Although the radioactive waste off-gas rates from BWRs are of the order of 440 m^3/h before the recombiners, the waste off-gas rates from PWRs average only several cubic meters per hour or less. Many of the recent radioactive waste gas treatment systems are more typically depicted in Figure 46. This is a more versatile system than that shown in Figure 45 and involves the use of recombiners. In addition to removing the hydrogen before it is stored, the design also allows for processing of the nitrogen cover gas used during shutdown for storage and reuse during the next shutdown.

Some PWR radioactive waste gas treatment systems also use charcoal adsorbents for holdup and decay of the noble gases. (This is also the case for the CANDU reactor.)

System Performance

The estimated annual emissions and performance of several radioactive waste gas streams and treatment systems for a typical 1000-MWe PWR are shown in Table 19.

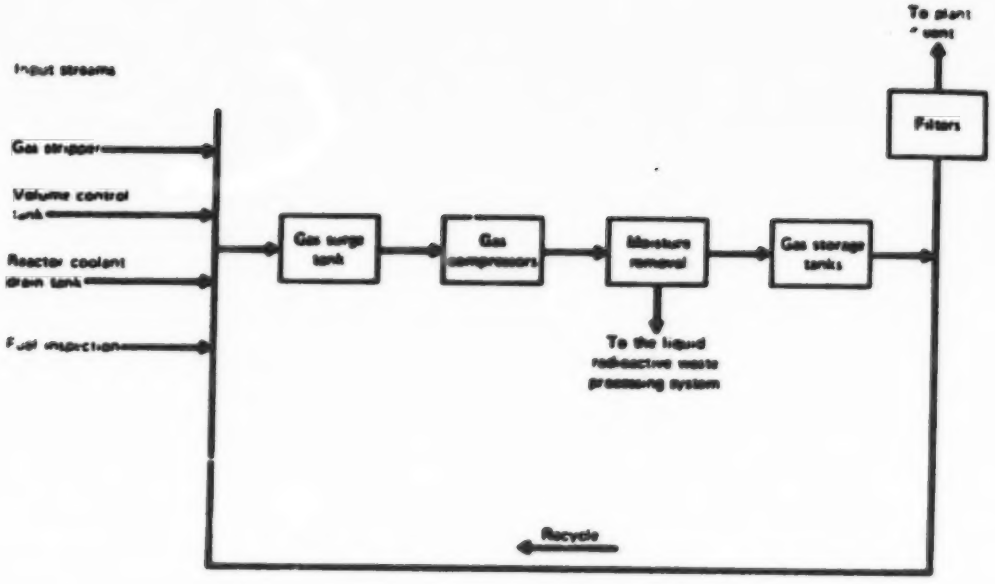

FIGURE 45 TYPICAL PWR TANK STORAGE SYSTEM FOR WASTE GAS (25)

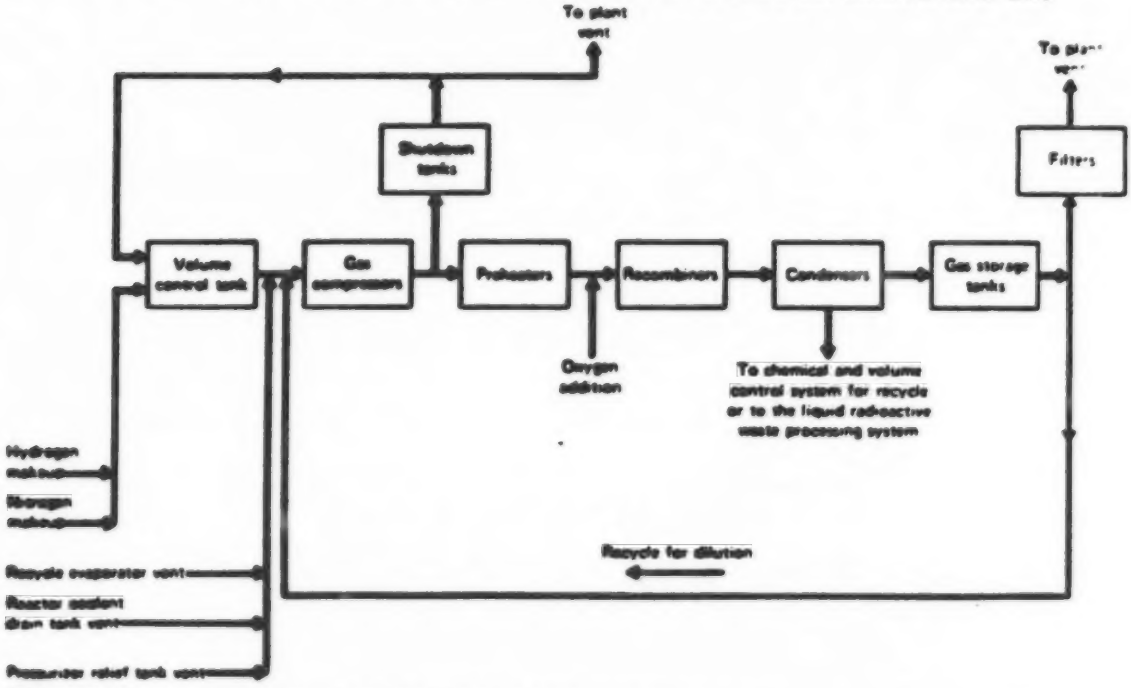

FIGURE 46 TYPICAL PWR VOLUME REDUCTION EXTENDED-STORAGE SYSTEM FOR USE WITH VOLUME CONTROL TANK STRIPPING (25)

TABLE 19 EFFECTIVENESS OF WASTE GAS TREATMENT SYSTEMS FOR NOBLE GASES IN 1000-MWe PWRs (25)

Source of release	Annual release, Ci
Waste gas treatment systems	
Untreated	316 000*
Charcoal delay (72-d delay for xenon; 4-d delay for krypton)	329*
Pressurized storage	
60 d	412*
90 d	306*
Estimated from other sources†	
Containment building	88†
Auxiliary building	10†

* Of this amount, ~300 Ci is ^{85}Kr.
† Based on full letdown of 0.00473 m^3/s (75 gpm).

All these control systems are cabable of reducing the noble gas emissions from PWRs by 99%, and some approach 99.9% reduction. Further reduction of emissions would necessitate the removal and storage of ^{85}Kr.

The status of radioactive waste off-gas treatment system technology for controlling noble gases in LWR effluents has been developed to the point where there appears to be little room or incentive for improvement. Techniques that are more cost effective and more acceptable from a safety viewpoint may be developed and applied, but improved performance does not appear to be needed. However, all these systems are based on holdup or delay and appear to be inadequate for handling larger off-gas flows that may result from accidents involving abnormally high emissions of noble gases from the fuel into the coolant systems.

5.0 Tritium Removal System

It is proposed to feed heavy water from the vapour recovery systems into the tritium removal system (TRS). In such a system currently being designed for application to CANDU stations, tritium is removed from heavy water in two steps, as indicated in Figure 47A (10). In the first step, the tritium is transferred from heavy water by Vapour

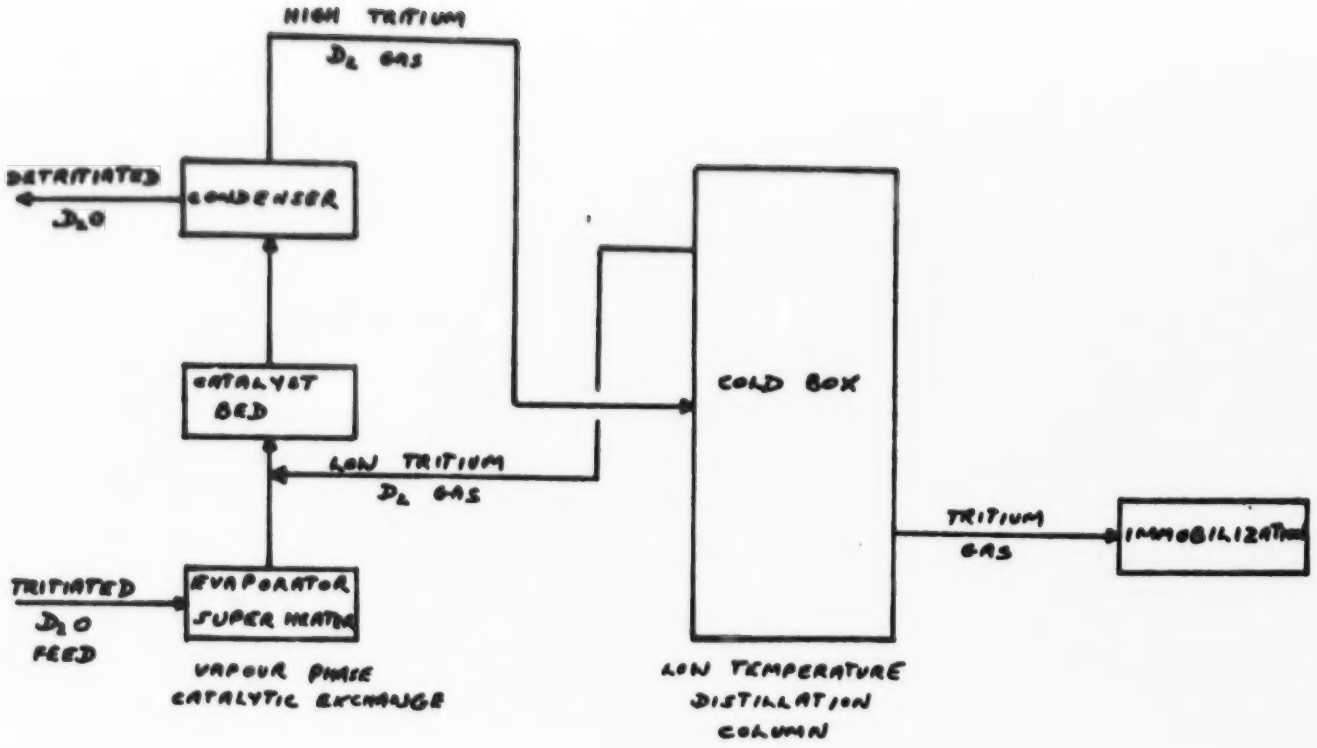

FIGURE 47A TRITIUM REMOVAL SYSTEM COMPONENTS (10)

Phase Catalytic Exchange (VPCE) with deuterium gas. The heavy water is evaporated, superheated to 200°C and mixed with deuterium gas before passing over à catalyst bed, where the following exchange reaction transfers tritium from the heavy water vapour to the deuterium gas.

$$DTO + D_2 \qquad D_2O + DT$$

In the second step, the D_2 and DT are cooled to -250°C and tritium is separated from deuterium by cryogenic distillation.

Figure 47B shows a more detailed schematic of the process. Further details are provided in ref. 10.

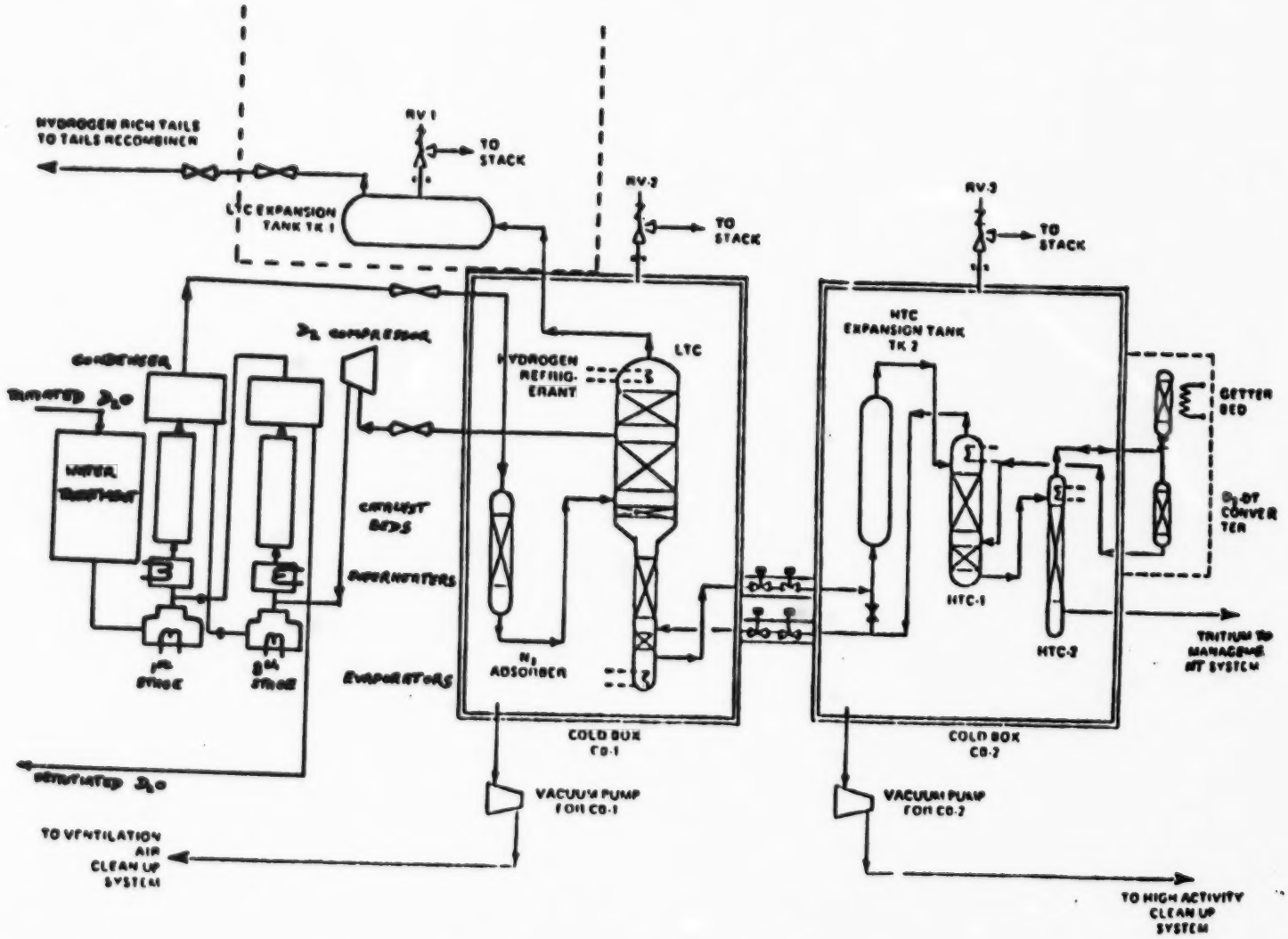

FIGURE 47B TRITIUM REMOVAL SYSTEM SCHEMATIC (10)

REFERENCES

1. Nuclear Energy Agency, "Objectives, Concepts and Strategies for the Management of Radioactive Waste Arising from Nuclear Power Programmes", OECD, Paris (1977).

2. Ontario Hydro, "Radiation Protection Training Course No. 1", Section 1-2-12.

3. Chemical Rubber Company (CRC), "Handbook of Chemistry and Physics." Editors West R.C. and Astle, M.J., 63rd Edition, 1982-1983.

4. Atomic Energy of Canada Ltd., "Recognition of Water Treatment problems in Thermal and Nuclear Power Stations, Control of Radioactive Build-Up, Hemmings, R.L. April 1973.

5. National Research Council Canada, "Radiation in the Canadian Aquatic Environment", NRCC No. 19250, 1983.

6. Ontario Hydro, "Quarterly Technical Reports", Nuclear Generation Division, from 1977 to 1982.

7. Guest, A., Maruska, R., "Monitoring of Airborne C-14 Emissions from Ontario Hydro Nuclear Generating Stations", A Presentation to the CRPA Conference (May 3 to May 5, 1983) Toronto.

8. Maan, M.A., Anyas-Weiss, N and Loritz, F., "A Study of Tritium Control in CANDU Nuclear Power Stations", American Nuclear Society. National Topical Meeting, Dayton, Ohio, 1980.

9. Carter, T.J., "Radioactive Waste Management Practices at a Large Canadian Electrical Utility, "IAE-SR-57/1, Seminar on the Management of Radioactive Waste from Nuclear Power Plants, Karlsruhe, oct. 1981.

10. Finlay, P.G., Christensen, R., Muir, J.D. and Draper, D.W., "Radioactive Liquid Wastes from Steam-Electric Generating Stations", Draft No. 3, Discussion Paper Prepared for Working Group 5 (2)" Radioactivity, Task Force on Steam-Electric Power Generation", EPS, Environment Canada, 1982.

11. Eichholz, G.G., "Environmental Aspects of Nuclear Power", Ann Arbor Science Publishers Inc., 1976.

12. Marois, H., Wilson, R.J., "Environmental Monitoring and Control of Radioactive Materials Released from Quebec and Ontario Hydro Nuclear Power Stations". CNA-74-606, 1974.

13. Kabat, M.J., "Experience with Gaseous Fission Products and Radioiodines at Ontario Hydro Nuclear Power Stations", paper presented at the 17th Annual Meeting of the Health Physics Society, Las Vegas, Nevada, 1972.

14. Watson, D.A. et al., "Gaseous Effluent Monitoring in Ontario Hydro Heavy Water Power Reactors, Monitoring of Radioactive Effluents, Proc. of the NEA Seminar, Karlsruhe, 1974.

15. Wong, K.Y., "Basis for the Derived Limits for the Release of Radionuclides in Gaseous and Liquid Effluents from Ontario Hydro's Nuclear Stations", Ontario Hydro Health Physics Department Report H.P.D.-73-6, Rev. 1 Aug. 1974.

16. London Nuclear Ltd., "Volume Reduction of Resin Wastes", Contract No. 000G157 Report prepared for the Canadian Electrical Association, July 1981.

17. Beninson, D., Gonzalez, A.J., "Application of the Dose Limitation System to the Control of Carbon-14 Releases from Heavy-Water-Moderated Reactors", presented at IAEA Symposium on the Dose Limitation System in the Nuclear Fuel Cycle and in Radiation Protection, Madrid, Oct. 1981.

18. Kabat, M.J., "Monitoring and Removal of Gaseous Carbon-14 Species", Ont. Hydro, 15th DOE Nuclear Air Cleaning Conference, Jan.-Feb. 1979.

19. Kabat, M.J., "Recent Developments in Gaseous Effluent Monitoring in Ontario Hydro, International Atomic Energy Agency, IAEA-SM-217/54, Vienna, 1978.

20. Jones, R.D. et al. "Snow and Tree Sap Sampling for Tritium near the Bruce Nuclear Power Development, 1982", Phytotoxicology Section, Air Resourcs Branch, Ontario Ministry of the Environment.

21. Kabat, M.J., "Selective Sampling of Hypoiodous Acid", 14th AEC Air Cleaning Conference, Sun Valley, Idaho, 1976 Proceedings.

22. Kabat, M.J., "Canadian Nuclear Air Cleaning Standards and their Application in Ontario Hydro", IAEA Seminar, "The Testing and Operation of Off-gas Cleaning Systems at Nuclear Facilities", Karlsruhe, FRG; 3-7 May 1982.

23. Neil, B.C.J., Pilar, G.J., "Carbon-14 in Biota from the Vicinity of Ontario Hydro Nuclear Generating Stations", Paper presented at 4th Annual Meeting of Canadian Radiation Protection Association, Toronto, May 4, 1983.

24. Burchsted, C.A., Fuller, A.B. "Design, Construction and Testing of High-Efficiency Air Filtration Systems for Nuclear Application". Report ORNL-NSIC-65, Oak Ridge National Laboratory (1970).

25. Pence, D.T., "Critical Review of Noble Gas Recovery and Treatment Systems", Nuclear Safety, Vol. 22, No. 6, Nov-Dec 1981.

APPENDIX A
NUCLEAR REACTIONS AND RADIONUCLIDE DATA

TABLE A-1 PARTIAL LIST OF NUCLEAR REACTIONS IN A CANDU SYSTEM

1. Natural Uranium Fuel

   - 99.3% U-238 (Fertile)
   - 0.7% U-235 (Fissile)

2. Typical Fission Reactions

   92-U-235 + o-n-1 → 38-Sr-94 + 54-Xe-140 + (2) o-n-1 + γ
   (Strontium and xenon)
   92-U-235 + o-n-1 → 39-Y-97 + 53-I-136 + (3) o-n-1 + γ

   (Fertilization of U-238)

   92-U-238 + o-n-1 → 92-U-239 → 93-Np-239 + β^- → 94-Pu-239 + β^-

3. Typical Fission Decay Chain Reactions

   38-Sr-94 → 39-Y-94 + β^- + γ
   → 40-Zr-94 + β^- + γ
   54-Xe-140 → 55-Cs-140 + β^- + γ
   → 56-Ba-140 + β^- + γ
   → 57-La-140 + β^- + γ
   → 58-Ce-140 + β^- + γ

   (Note: Over 200 fission products have been identified.)

4. Typical Fission Product Gases which would escape on fuel sheath failure:

   Xe (Xenon)
   Kr (Krypton)
   I (Iodine)
   Ru (Rubidium)
   Ce (Cesium)

5. Typical Fission Gases Decay Chain Reactions

   53-I-135 → 54-Xe-135 + β^- + γ
   → 55-Cs-135 + β^- + γ
   → 56-Ba-135 + β^- + γ
   36-Kr-88 → 37-RB-88 + β^- + γ
   → 38-Sr-88 + β^- + γ

6. Typical material Corrosion Products Activation Reactions

   27-Co-59 + o-n-1 → 27-Co-60
   27-Ni-58 + o-n-1 → 27-Co-58
   24-Cr-50 + o-n-1 → 24-Cr-51

7. **Typical Fluid Activation Reactions**

1-H-2 + o-n-1 (Deuterium)	→	1-H-3 + γ (Tritium)
8-O-16 + o-n-1	→	7-N-16 + 1-p-1
8-O-18 + o-n-1	→	8-O-19 + γ
8-O-16 8-O-17 8-O-18 6-C-13 *7-N-14 *7-N-15	→	6-C-14
18-Ar-40 o-n-1	→	18-Ar-14 + γ

*Also from Nitrogen impurities in fuel

TABLE A-2 SIGNIFICANT FISSION PRODUCTS FROM CANDU REACTORS (2)

Element	Symbol	Contribution to total activity, %					Half-Life
		1 day	10 days	30 days	90 days	1 year	
Strontium	Sr^{89}		2.8	6.7	10.5	1.7	53d
Strontium	Sr^{91}	6.7					7.7h
Strontium	Sr^{90}						28y
Yttrium	Y^{90}					1.8	64
Yttrium	Y^{91}		3.4	7.6	12.5	3.9	60d
Yttrium	Y^{92}	4.2					3.6h
Yttrium	Y^{93}	7.6					10h
Zirconium	Zr^{95}		3.7	8.2	14.7	7.3	65d
Zirconium	Zr^{97}	9.0					17h
Niobium	Nb^{95}			4.1	18.2	15	35d
Niobium	Nb^{97}	9.6					72m
Molybdenum	Mo^{99}	4.6	6.8				66h
Ruthenium	Ru^{103}		2.65	5.7	7.2		40d
Ruthenium	Ru^{106}					2.4	1.0y
Rhodium	Rh^{106}						30s
Rhodium	Rh^{103m}		2.55	5.5	7.0		57m
Rhodium	Rh^{105}	1.35					36h
Tellurium	Te^{132}	2.6	5.1				78h
Iodine	I^{131}		6.8	3.7			8.1d
Iodine	I^{132}	2.7	5.3				7.3h
Iodine	I^{133}	7.3					21h
Iodine	I^{135}	4.7					6.7h
Xenon	Xe^{133}	1.23	11.4	2.6			5.3
Xenon	Xe^{135}	12.5					9.2h
Cesium	Cs^{137}						30y
Barium	Ba^{137m}					1.5	2.6m
Barium	Ba^{140}	1.25	10.6	10.8	1.6		12.8d
Lanthanum	La^{140}		12.0	12.5	2.4		40h

TABLE A-2 SIGNIFICANT FISSION PRODUCTS FROM CANDU REACTORS (2) (Cont'd)

Element	Symbol	Contribution to total activity, %					Half-Life
		1 day	10 days	30 days	90 days	1 year	
Lanthanum	La^{141}	1.4					3.9h
Cerium	Ce^{141}		6.3	11.2	8.5		32.5d
Cerium	Ce^{143}	6.8					33h
Cerium	Ce^{144}						285d
Praseo-dymium	Pr^{144}			2.0	6.0	27	17.3m
Praseo-dymium	Pr^{143}		10.0	11.2	1.9		13.7d
Neodymium	Nd^{147}		4.8	4.1			11.1d
Promethium	Pm^{147}					5.7	2.7y
Promethium	Pm^{149}	1.45					53h

TABLE A-3 SIGNIFICANT ACTIVATED CORROSION PRODUCTS FROM CANDU REACTOR CIRCUITS (10)

Parent	Nuclide	Half Life
cobalt-59	cobalt-60	5.26 years
iron-54	manganese-54	300 days
nickel-58	cobalt-58	71 days
iron-58	iron-59	45 days
chromium-50	chromium-51	28 days

TABLE 4 SIGNIFICANT SYSTEM FLUID ACTIVATED PRODUCTS FROM CANDU REACTOR CIRCUITS

- Dissolved and Suspended Fuel Fission Products (See Table 2)
- Dissolved and Suspended System Fluid Activated Products (See Table 3)
- Oxides of Tritium

Parent	Nuclide	Half Life	Sp. Activity
deuterium-2	tritium-3	12.33y	9.64×10^3 Ci/g

APPENDIX B
TRITIUM-IN-AIR-MONITOR

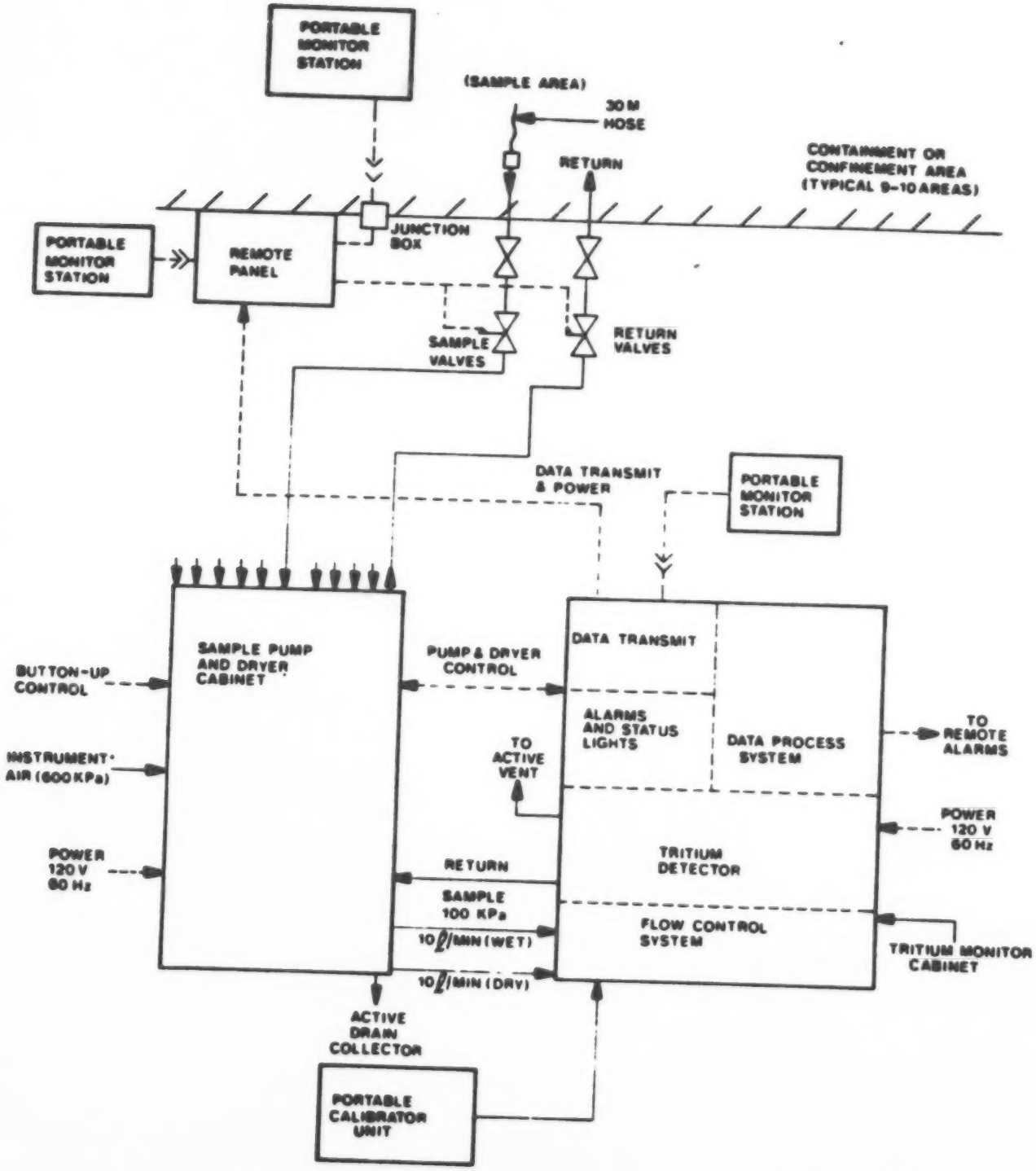

FIGURE B-1 BLOCK DIAGRAM OF THE TRITIUM-IN-AIR MONITORING SYSTEM

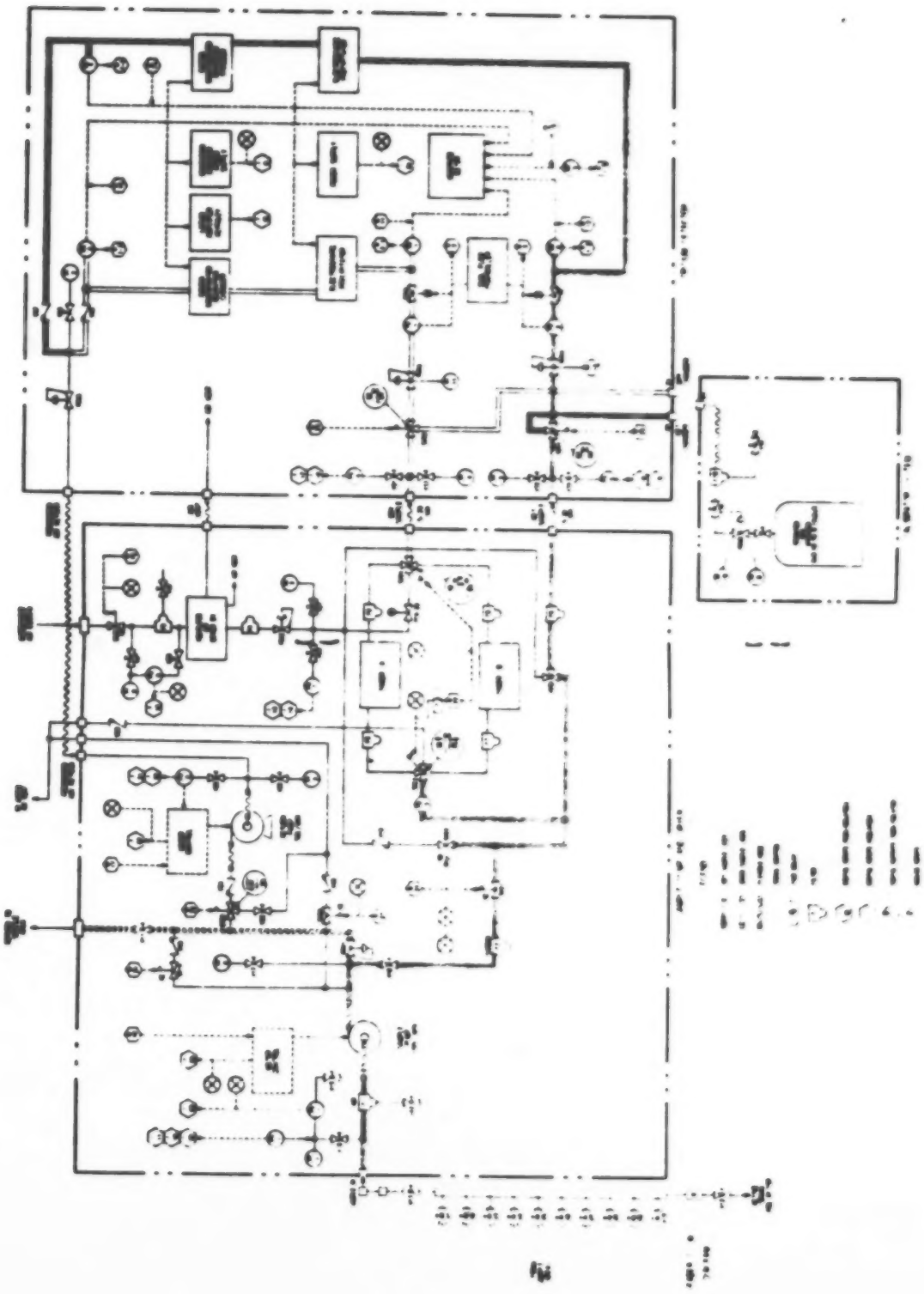

FIGURE B-2 PNEUMATIC LAYOUT FOR TRITIUM-IN-AIR MONITOR

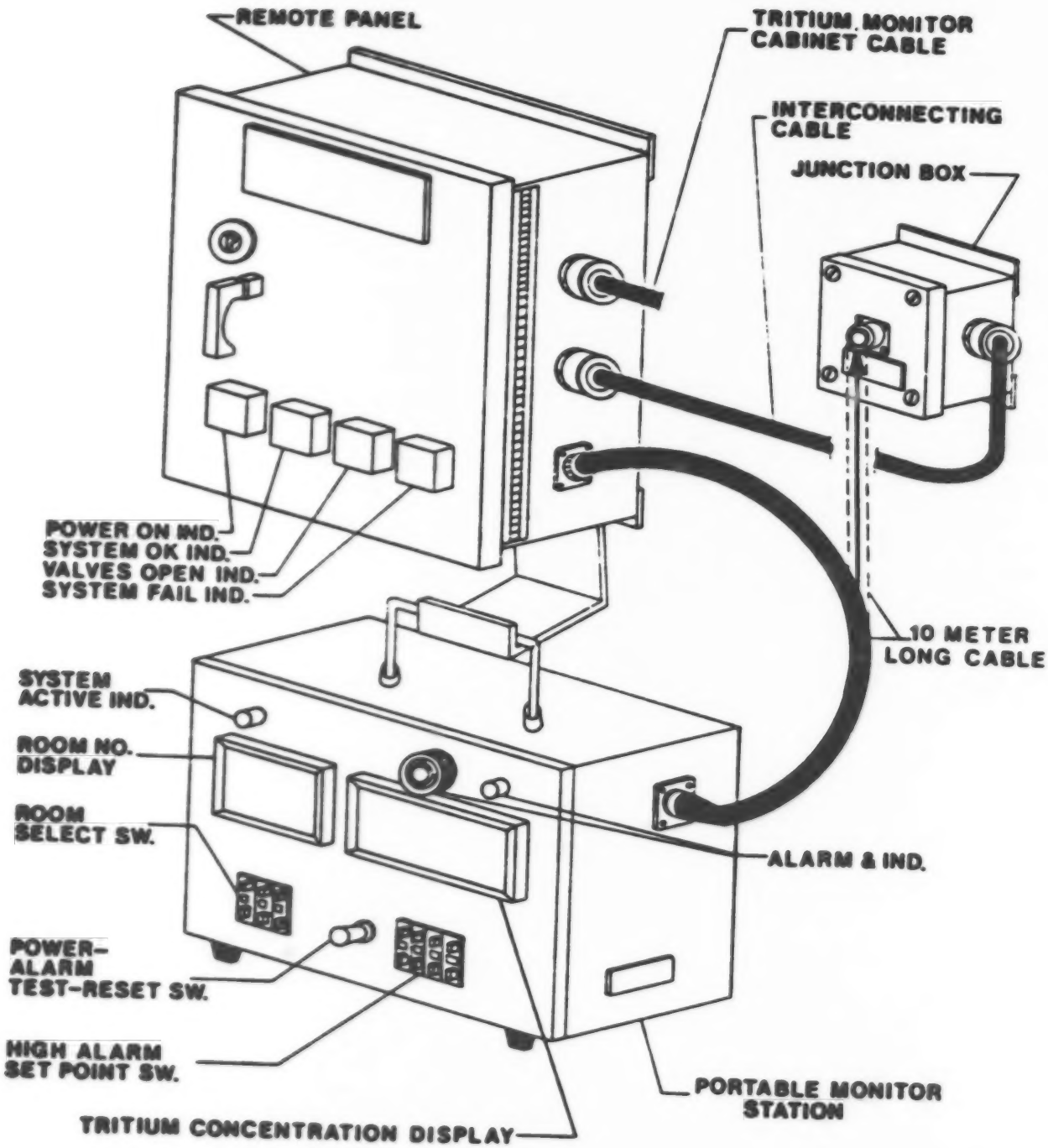

FIGURE B-3 REMOTE PANEL, PORTABLE MONITOR AND JUNCTION BOX LAYOUT